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Differential evanescent light intensity evaluation of a-Se nanostructure

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Using the technique of capturing the evanescent light leaking image, named recently Differential Evanescent Light Intensity (DELI), which yields information about the photodeposited nanostructures in the deposited zone we evaluated the morphology of ultra-thin a-Se photodeposited nano-structures deposited directly on glass substrates serving as waveguides by high intensity Xenon irradiation lamp in the visible spectrum. Deposition fluences of about $F \approx 300 J/cm^2$ were enough to produce layers up to about $170 nm$ thickness, similar to values needed for CW Ar^+ ion laser PD deposition at λ = 498 nm reported in previous investigations. The deposited particles observed on the substrates have diameters typically in the range of 100-300 nm as obtained by other microscopy methods. The morphology of the nano-structures observed by DELI during this work conditions consist of a random array of individual particles adsorbed onto the surface. The deposited material morphological profile was observed for fluences in excess of $F > F_{th} \sim 25$ J/cm^2 for the particular experimental conditions of this work for a-Se. The highest merits found for the DELI technique are the fast and ease of measurements, better z-resolution than SEM and capability of large areas profiling and mean thickness measurements.

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1. Introduction

Photodeposition (PD) from solutions can be used for realizing various thin film patterns, of sub-microscopic thicknesses i.e., 5-500 (nm) to produce various spatially distributed components for optical applications such as described in [1]. Various patterns can be written in one step on solid interfaces which are in contact with the solution by laser scanning or projection and mask imaging. The rate of the process for various materials depends on the laser wavelength, fluence and temperature of the photoreactor.

During PD [2] nanometer particles appear on the irradiated zones of any transparent substrates, such as glass used in this investigation. In this work, CW Photodeposition from a-Se colloid solutions onto glass substrates with a Xenon UV-Visible lamp has been employed.

The sensitive evanescent method named differential-evanescent light intensity (DELI) for surface nanometer particles investigation was used to evaluate and estimate quantitatively the deposited nanolayers profiles. This waveguide differential-evanescent light extraction (DELI) technique has the advantage that the propagating beam field does not interfere with the electromagnetic field extracted perpendicular to the substrate surface. Evanescent wave light illumination is used recently as a method for excitation and detection of scattering and fluorescent objects near the surface by thin samples. This is because the technique gives a dark background, providing good spatial resolution for small objects near the surface structures in comparison with other microscopy techniques [3].

This nanometer profilometry evaluation sensitive method was used by us in a previous work on PD of a-Se obtained by PLD by an ArF UV excimer laser allowing us to evaluate 5-20 nanometer thick deposited profiles, and their growth rate [4].

2. Experimental procedure of CW photodeposition

The deposition of the nanometric a-Se layers was performed as follows: The light beam from a UV-visible spectrum CW Xenon discharge lamp was shaped into a circular shape of homogenous light irradiating an area of about 1 cm^2, at the substrate-colloidal a-Selenium solution interface. The substrates were microscope slides made of Borosilicate glass with dimensions of 76x25x1 mm onto which the a-Se nanolayers were deposited. The solution was a metastable hydrosol which deposits a-Se on the glass interface upon irradiation with Visible Short Wavelength light (Blue Spectrum). The total irradiation intensity of the lamp, after the circular aperture, was about 1.6 watts/cm^2 in the visible range from 390-600 nm. The lamp was a Dr. Höhnle Co. fiber illuminator, type "Blue Point" (Xenon Lamp). The light spectrum shape is shown in Fig.1. The UV spectrum light in the wavelength zone below 390 nm was filtered by the Borosilicate glass.

To obtain a-Se deposited material with various nanometric thicknesses, an electronic shutter was used to adjust the cumulative irradiation dose in time, as required.

Thus, the beam fluence at the substrate-solution interface was varied in the range 10-290 (J/cm^2) by changing the deposition time.

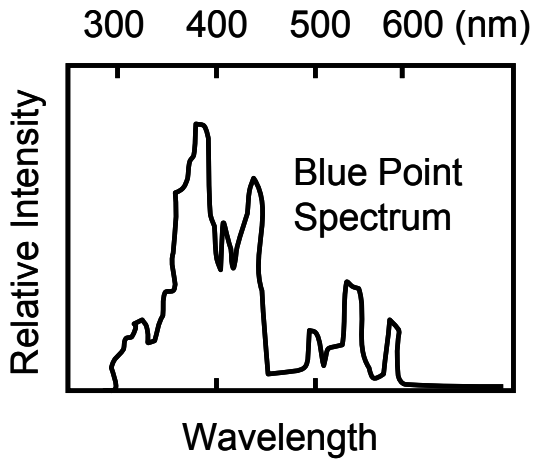

Fig.1. The spectrum of Dr. Höhnle Co. fiber illuminator, type "Blue Point" (Xenon Lamp) used for photodepositing a-Se layers. The intensity of the light in the spectral zone of 390-600 nm was 1.6 Watt/cm^2.

3. a-Se Nanometer films profile evaluation by DELI

The profiles of thin films of the a-Se deposited nanolayers were observed directly on the glass substrates by the Differential Evanescent Light Intensity (DELI) method [5]. To observe the morphology we captured 2D images at several zooming powers of a microscope equipped with a CCD camera, down to the range of well resolved x-y areas.

Our technique of Differential Evanescent Light Imaging (DELI) used in this investigation is a version of the far field technique developed to achieve a high resolution for nanometer features in the z-direction, $i \cdot e$, into the depth direction of the surface samples. The technique uses basically the phenomenon of Total Internal Reflection (TIR) [6,7] where evanescent waves arise.

This technique has the advantage that the far field image does not include direct transmitted or reflected light. In fact we rely on the "evanescent light extraction power" of the nanoparticles and nanolayers on the substrate which serves as optical waveguide. We usually capture the evanescent image in the far field since we are interested in large areas structural investigation but proximity observation is also possible. The TIR phenomenon occurs when light passes from an optically denser medium n_1 which is the waveguide, into a less dense medium n_2. The evanescent light intensity I_{ew} field in z- direction is described after a regular refraction incidence at the interface by:

$$I_{ew}(z) \sim I_0 \cdot e^{-\frac{2 \cdot z}{d}} \tag{1}$$

Where I_0 is the initial light intensity in the waveguide , and the characteristic penetration depth is defined by:

$$d = \frac{\lambda_{vac}}{2 \cdot \pi \cdot \sqrt{\left(n_2^2 \cdot \sin^2 \theta_i - n_1^2\right)}} \quad (nm) \tag{2}$$

here λ_{vac} is the free space wavelength of the light beam used.

The DELI technique uses the optical light intensity pattern captured by the deposited particles at the substrate/interface from a visible light beam propagating through the glass substrate which serves as light waveguide.

The light source used was a GE-Thorn, Quartz Halogen lamp; model DDL, with a fiber illuminator, type Volpi AG, supplying an integrated light intensity of about I_0 = 3.75 W/ cm^2 in the visible region of 400-800 nm at the fiber tip. The optical image of the evanescent emanating light from the waveguide was captured by a zooming microscope equipped with CCD camera, with a peak wavelength response at 555 nm, and then integrated over the whole area into an effective gray level Integrated Optical Density (IOD) which is approximately proportional to the nano-layers thickness. Using a simplified formalism we related the IOD linearly to the nanometer thickness of layers as shown later.

The evanescent light intensity extraction by the nanolayer of thickness *h* in the near as well as in the far field zones from the optical waveguide can be modelled by the following equation:

$$I_{ew}(h) = \int_0^h K(z) \cdot I_0 \cdot e^{-\frac{2 \cdot z}{d}} dz \tag{3}$$

which incorporates the light extraction efficiency, $K(z)$, due to the optical interactions prevailing at the interface between the nanoparticles and the light in the waveguide below.

Assuming a simple formal dependence for the light extraction efficiency by the particles in terms of the nanoparticle's concentration, *M(x,y,z)* (*cm^{-3}*) and the evanescent scattering cross section, σ_{scatt} (*x,y,z),* (*cm^2*):

$$K(z) = M(x, y, z) \cdot \sigma_{scatt}(x, y, z) \tag{4}$$

By modelling the M and σ_{scatt} dependences we can obtain from eqn. (3) the near/ far field image emerged from the nanolayer and hence obtain the optical response of the CCD camera which is proportional to the intensity of light extracted by the nanolayer.

For constant M and σ_{scatt} we get the following estimation for the relative intensity changes in the CCD

image obtained from the evanescent field extraction by the nanolayer of thickness h:

$$\eta = \frac{I_{ew}(h)}{I_0} = \frac{1}{2} Kd \cdot (1 - e^{-\delta h}) \quad \text{with:} \quad K = \sigma_{scatt} \cdot M \quad \text{and} \quad \delta^{-1} = d/2 \tag{5}$$

where $I_{ew}(h)$ is the intensity of light extracted by the nanolayer of thickness $h(x,y)$, and d is the penetration depth from eqn. (3) .

We obtain thus the layer thickness profile from the Integrated Optical Density measurements and assuming that $\frac{IOD_1}{IOD_2} \approx \frac{\eta_1}{\eta_2}$.

For the simplest mathematical case of very thin films $\delta \cdot h << 1$ we obtain from (5) a linear proportionality between nanolayer thickness and the normalized intensity, so as for two different thicknesses the Integrated Optical Density measured gives an estimation of the nanolayer thickness:

$$\frac{IOD_1}{IOD_2} \approx \frac{\eta_1}{\eta_2} = \frac{(1 - e^{-\delta h_1})}{(1 - e^{-\delta h_2})} \approx \frac{h_1}{h_2} \tag{6}$$

Calibrating for h_2 one get a convenient method of estimation for any other thicknesses h_1 from (6). If an absolute thickness profile is required, h_2 must be measured by independent techniques such as Spectrometric, SEM, AFM or mechanical nanoprofilometer and then calibrating the IOD values accordingly.

4. DELI results of the morphology of the nanolayers

In Fig.2 we present typical images of a-Se zones, deposited with a fluence of 192 J/cm^2 as obtained by the Differential Evanescent Light Intensity (DELI) method. Typical DELI 3D perspective images of an a-Se deposited area of 400 x 400 μm is shown in Fig. 2 a). Then expanded 2D- and 3D-views at higher magnifications available by DELI showing particles down to about 125 nm diameter.

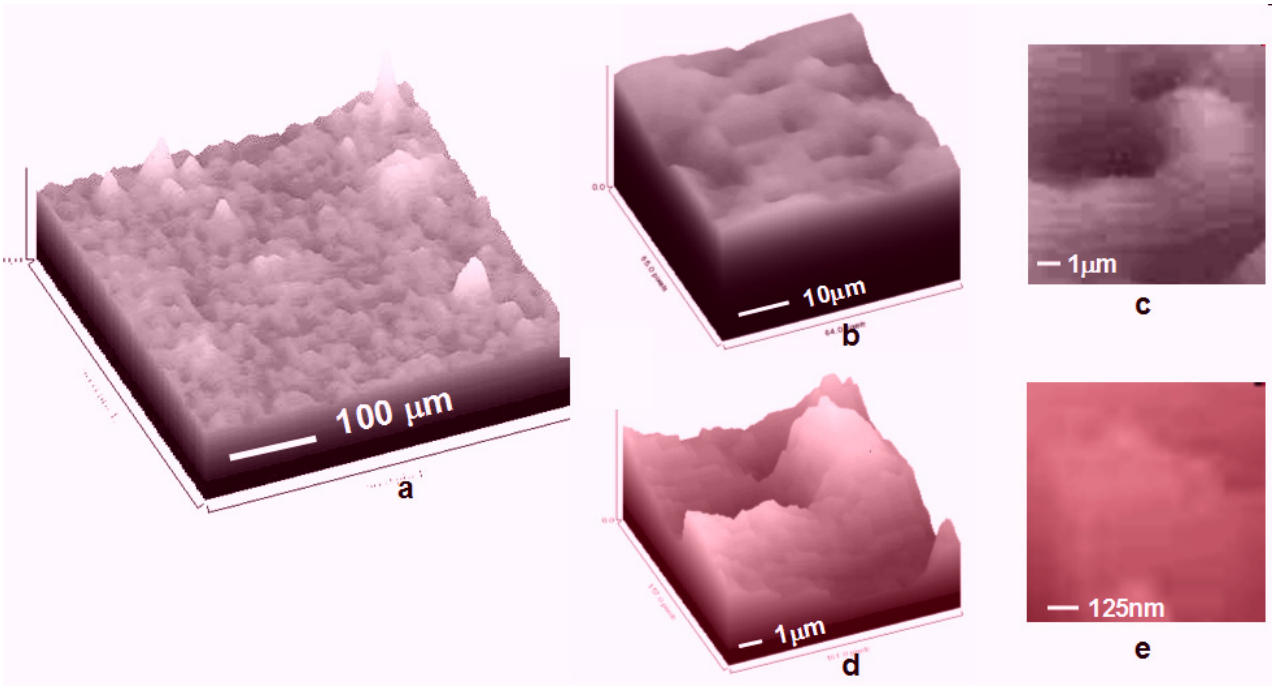

Fig.2 a). Typical DELI 3D image of an a-Se typical deposited area; b). Magnified area of a.); c). 2D-view of a chain of 4 particles; d). 3D-view of fig.c).; e). 2D-view at the highest magnification available, showing the substructure of a particle.

From the DELI calculations based on Fig. 2 we deduced a lateral resolution of 1 μm, and in the z-direction of about 100 nm. As can be observed, the morphology at this resolving power gives for the sample shown, a typical surface structure of isolated spherical particles and interconnected spheroids. For the thicker samples we observed similar forms but with a higher connectivity and merging between the spheroids. It can also be seen that they are also more bulky than the thinner samples.

Thus using the evanescent method for capturing the surface details optically in conjunction with computer image processing methods, the surface morphology and depth z- profiles of nanolayer samples can be obtained in 2D or 3D images in a convenient and with high contrast by the evanescent field.

Using this technique we obtained the Integrated Optical Density and calibrated absolute thicknesses of the deposited zones for the whole fluence deposition range of 10-290 (J/cm^2) see Fig. 3. As seen, the IOD is proportional approximately to the nano-layers profile thickness.

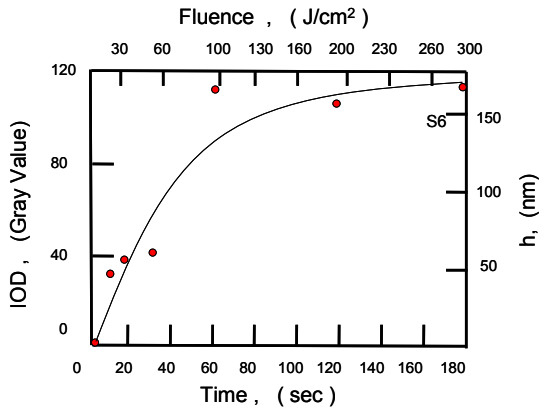

Fig. 3. IOD of several samples on substrate S6 as a function of deposition time, corresponding to a deposition fluence range of 10-290 (J/cm^2). The maximum thickness on the curve is about ~ 170 nm as obtained by calibration from AFM measurements. The fluence corresponding to the second point on the curve is ~ 10 (J/cm^2).

5. Discussion

This investigation showed that the profiles of the a-Se colloid adsorption in the irradiated zone for the fluence range of 10-290 J/cm^2 can be observed by DELI. The smallest deposited layer thickness was about 45 (nm) and the highest, typically about 170 nm. The final deposited material structure seen is inhomogeneous across the deposited zones since the depositing light source intensity profile was not entirely homogenous.

The film thickness grows monotonically up to about 170 nm thickness under the particular experiments conditions used. From Fig. 3 we can also conclude that the fluence threshold value for photodeposition under these conditions is less than 25 J/cm^2. Sub-particles of 125 (nm) diameter could be observed in the DELI images.

6. Conclusions

The motivation for investigating the a-Se nanolayers deposited by photodeposition with CW lamp method was to analyze the process suitability for controlled nanometer thickness deposition and nanometer profile evaluation by the simpler and convenient differential-evanescent light extraction technique from an optical waveguide. We have realized that it is possible to control the deposited thin films thickness by the light dose mainly, achieving controllable nano-layers in the range of 45-170 nm. The foremost specific advantages of the DELI technique used found were is its simplicity and non-destructive features as compared to other more expensive evaluation methods such as SEM and AFM, and suitable especially for large areas evaluation of nano thickness films profiles.

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