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Observation and analysis of self-organized surface grain structures in silica films under nonepitaxial growth mode

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Abstract

Silica films under present reactive electron beam deposition conditions have depicted a novel self-organized surface grain structures when probed through atomic force microscopy, 2D fast Fourier transform and glancing incidence X-ray diffraction techniques. The formation of such ordered surface grain structures is observed to be strongly correlated to the nucleation and growth process of the silica films. However, the nature of the substrate (amorphous or crystalline) and multilayer geometries have influenced the shapes, sizes and abundances in the grain structures and the ordering. The strain mediation of such ordered structures when buried under polycrystalline layers like Gd_2O_3 have shown to influence both the grain size as well as roughness. A variety of grain structure evolutions and morphological changes in silica layers were noticed in different multilayer geometries. It is, hence, inferred that by appropriately using combinations of these materials, it is possible to have a control over the multilayer morphology and grain structures, which is a very relevant factor in developing precision ultraviolet laser coatings.

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Keywords: Self-organization; Morphology; Atomic force microscopy; Ordered grain structures; Fast Fourier transform; Optical thin film; Glancing incidence X-ray diffraction

1. Introduction

Silicon dioxide, more popularly known as silica, has always generated great interest in coating and interface layer technologies because of its excellent physical and chemical properties. As a low refractive index material, silica (SiO_2) is the only reliable refractory oxide thin film material to meet the wide range of spectral requirements in this wavelength region [1]. SiO_2 films are known for their high band gap and multilayer compatibility with a large number of refractory oxide optical coating materials. It is possible to grow this

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material in various structural forms ranging from amorphous to nano- and mesostructures. It demonstrates several optomechanical novelties under each structural form. There has been great interest in investigating the structure of thin SiO_2 films and buried SiO_2/Si interfaces for applications relating to metal-oxide semiconductor devices [2]. Several well-established approaches have been employed in growing such films that include oxidation of silicon at elevated temperatures, pyrolytic decomposition, plasma-enhanced chemical vapor deposition, reactive sputtering, radio frequency magnetron sputtering, etc. [3]. Film deposition and post-deposition process conditions strongly affect the film microstructure and consequently, some of their microscopic properties. The novelty of our present work lies in the observation of self-organized surface grain structures of SiO_2 films grown nonepitaxially, i.e., under reactive electron beam deposition technique. There are some interesting morphological, as well as ordered structure symmetry outcomes which were observed in these grain structures. The origin and evolutions in the shapes and sizes in the ordered grain structures has a great similarity to SiGe/Si and InAs/GaAs systems grown under strain fields. Although our processing conditions are entirely different, the ordered three-dimensional (3D)-island growth structures in SiO_2 films, both in single and multilayer geometry, very much resemble the above-mentioned epitaxial and heteroepitaxial semiconductor films and multilayer systems [4]. In the present work, the well-established strain-mediation theory usually employed for such microstructural evolutions have been adopted to explain the possible origin of the ordered SiO_2 grain structures in our multilayer geometries [4,5].

2. Structural ordering and self-organization process

Ordered microstructure in SiO_2 films has recently drawn special attention amongst various researchers, especially with relationship to the semiconductor device applications [6,7]. There have been a few reports on the direct evidence of epitaxial ordered oxide in thermally grown SiO_2 films. Munkholm et al. have observed that such an ordered structure of microcrystallites give rise to modulations of the crystal truncation rod (CTR) scattering [8]. The oxide ordering increases with the film thickness and the lack of higher-order reflections suggests that the crystallinity of the oxide is rather poor, with a large amount of static disorder [8]. Several novel epitaxial approaches have also been reported over the period to grow such ordered SiO_2 structures. Morphology and oxidation kinetics of SiO_2 structures have also drawn special attention both relating to basic understanding as well as technological applications of the self-organizations and their ordering processes [9]. Self-organization is a frequently encountered phenomenon in the epitaxial SiGe/Si systems [4,10,11]. Also, increasing number of experimental and theoretical studies have been devoted to self-organized surfaces during the last few years on varieties of semiconductor materials and processes [12–14]. It was proposed very early that these surfaces could serve as ideal templates for the growth of 1D or 2D arrays with nanometer scale dimensions.

Most commonly, self-assembly and self-organization are simply thin film growth processes obtained when a deposited material does not wet a surface [15]. The structures obtained might be islands, wires, networks, etc. ‘‘Assembly’’ versus ‘‘organization’’ is a semantic issue often discussed, not all researchers use the same definition. In a general perception, ‘‘self-assembly’’ is designated as the case, where atoms deposited on a surface aggregate spontaneously in structures, whereas, ‘‘self-organization’’ is the case where these structures display a well-ordered crystal-like positional order [16]. Therefore, self-organization is viewed as a special case of self-assembly. For most thin films with appropriate thickness and temperature, 3D growth can be favored by surface energy or parameter mismatch phenomena, resulting in the formation of structures. For epitaxial or heteroepitaxial systems, such organization results from an existing self-organized pattern on the surface acting as periodic nucleation centers. This self-organized pattern may also be induced by arrays of dislocations, surface reconstructions, adsorbates, etc.

3D self-organization is, however, not restricted to epitaxy only. The stacking of Co clusters with layers of amorphous alumina was reported to give rise to a super-FCC stacking [17]. As mentioned above, in most epitaxial systems and growth modes, the substrate plays a vital role in the initiation of the self-organization process. However, in nonepitaxial processes, the mechanism of self-organization has not been explored adequately to depict any convincing explanation. In these systems, the possible mechanism of formation of such an organization might be a direct interaction between the structures evolved during the nucleation and growth process. The experimental results usually point out to the more likely strain and stress-instabilities phenomena for the organized structures. In some experiments, corrugation correlations are noticed to be responsible for the progressive occurrence of order in nonepitaxial systems [17]. In this report, appearance of novel self-organized SiO_2 ordered surface grain structures and their contribution towards morphological evolution under single and multilayer geometry has been presented. For precision optical coating applications, such morphological information and analysis are most often essential to improve upon the performances of the multilayer devices.

3. Experiments

Under the present investigation, we have carried out some systematic experiments and analysis on post-growth microstructural analysis of SiO_2 films, using a multimode scanning probe and glancing incidence X-ray diffraction (GIXRD) techniques. The samples were deposited in a fully automatic thin film vacuum system "VERA-902" by adopting the reactive electron beam deposition technique. The depositions of the films were carried out using an 8 kW VTD electron beam gun with sweep and automatic emission controls. The film materials for SiO_2 and Gd_2O_3 were chosen from Cerac's batch number "S-1060" (purity 99.99%) and "G-1076" (purity 99.9%), respectively. The substrate temperature was maintained at 70 °C for both the films. The total pressure inside the chamber during the deposition process was maintained at 1×10^{-4} mbar through MKS mass flow controllers. The constituents of the gases present during the deposition were analyzed by a residual gas analyzer model; Pffeier's Prisma-200. One of the very noticeable effects is the presence of reasonable amount of residual water vapor in the chamber, whose partial pressure value was in the range of 1×10^{-6} mbar during the reactive deposition. The film thicknesses were monitored using both the Leybold's OMS-2000 OTM (optical thickness monitor), as well as Inficon's XTC/2 QCM (quartz crystal monitors). The typical rate of depositions was maintained at 1 nm/s for most of the films. The entire deposition process parameters such as substrate temperature, rate of deposition and total reacting gas pressure were monitored and controlled by a Siemen's industrial programmable logic controller with an appropriate frontsoftware. The film thicknesses were kept constant at quarter wave thickness at 248 nm (KrF laser) wavelength. This amounts to be approximately 31 nm for Gd_2O_3 and 41 nm for SiO_2 layers in terms of their physical thicknesses.

For atomic force microscopy (AFM) characterization, NT-MDT's solver P-47 H multimode ambient-based scanning probe system has been utilized. The cantilever used was a Si_3N_4 with a typical spring constant of 0.6 N/m and resonant frequency of 75 kHz. We have adopted the contact mode operation without any image filtering technique for the topographic measurements. For Fourier analysis, the built-in fast Fourier transform (FFT) module of the control software "NOVA-SPM" was employed to generate the mappings.

Films deposited by reactive electron beam process on various substrates (amorphous BK7 and crystalline silicon) and single or multilayer geometries have depicted a novel self-organized surface grain structures. The strain mediation effect of the self-organized grain structures on the morphology has been studied by employing several periodic SiO_2/Gd_2O_3 and SiO_2/TiO_2 systems starting from single to four layers on crystalline (Si), as well as amorphous (BK7, fused SiO_2) substrates. The reason for using gadolinia (Gd_2O_3) and titania (TiO_2) as associated layers with SiO_2 in the present experimental studies is that such combinations have a great potential in developing optical multilayers for various ultraviolet and visible laser applications [18–20]. Besides, it is also aimed to observe the strain mediation effects of polycrystalline Gd_2O_3 and TiO_2 in the structural ordering, as well as morphology in SiO_2 grain structures. The depositions of various single and multilayer films have been carried out using a process-controlled reactive electron beam deposition process in a fully automatic VERA-902 system and the details of these experiments can be found elsewhere [18]. In the present work, AFM and 2D FFT analysis techniques have been employed in probing, as well as analyzing the grain structures, their symmetry and ordering process. Thicknesses of single and multilayer films were decided keeping ultraviolet excimer laser-based applications in mind (physical thickness of Gd_2O_3 and SiO_2 are 31 and 41 nm, respectively) [20]. In the following sections, the thin film layer "H" corresponds to a high-index material, which is either Gd_2O_3 or TiO_2, and "L" corresponds to the SiO_2 films.

In order to support the AFM measurements, some of the samples were also characterized by the GIXRD technique. For GIXRD technique, a STOE X-ray diffractometer with CuKα source was used in the grazing incidence geometry for structural characterization of the films and multilayers. The presence of sharp, well-defined peaks clearly indicate the presence of organized grain structures in the SiO_2 films. These results also indicated the cubic nature of the gadolinia polycrystalline films.

4. Results and discussions

4.1. Results of the AFM measurements

Fig. 1(a) depicts the AFM topographic imaging of the single-layer SiO_2 film deposited on Si substrate. It shows the presence of SiO_2 microcrystalline grain structures grown in a reasonably ordered fashion. The self-assembled surface microcrystal-like grains depict a highly regular and aligned structure. The average (mean) and RMS roughness values for this sample are measured to be 4.90 and 6.21 nm, respectively over a scan area of ~$5 \times 5\,\mu m$. The FFT analysis result of this topography is presented in Fig. 1(b). This 2D-FFT spectrum distinctly shows a two-fold dominant symmetry at the center, whereas the outer diffuse pattern shows four-fold symmetry.

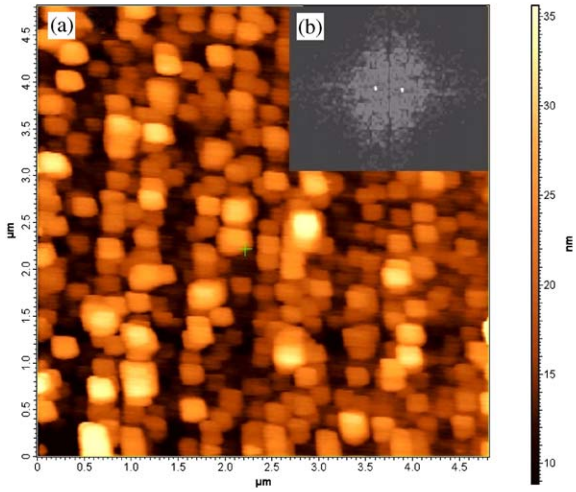

Fig. 1. (a) AFM's surface topographic image and (b) its FFT spectrum of single-layer SiO_2 films deposited on Si (1 1 1) substrate. The self-organized surface grain structures are distinctly evident in the measurement. The average roughness is 4.90 nm.

Similar SiO_2 films grown on BK7 (amorphous) substrate were also analyzed for their topographies. The measurement results are depicted in Fig. 2. It can be noticed from Fig. 2(a) that although there is a fair amount of orderliness in the grain structure, the density factor for the grains (i.e., total number of grains under the scan area of ~$5 \times 5\,\mu m$) has changed appreciably. The shapes are more of rectangular type compared to their counter parts on the Si substrate. The surface grain distributions appear more similar to microcrystallite-like structures in an amorphous matrix [21–23]. The diffraction peaks in the FFT spectra (Fig. 2(b)) are broadened and outer distributions form a diffuse ring structure indicating the presence of a strong isotropic component along with the ordered grain structures. From these two measurements, it is quite apparent that the formation of ordered grain structure in nonepitaxial systems is mostly dependent on the

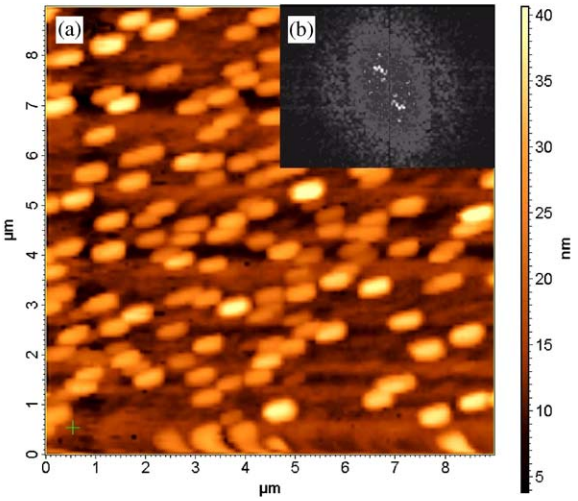

Fig. 2. (a) AFM's surface topographic and (b) its FFT spectrum of single-layer SiO_2 films deposited on BK7 glass substrate. The self-organization process appears to be somewhat different on an amorphous substrate. The average roughness is 6.05 nm.

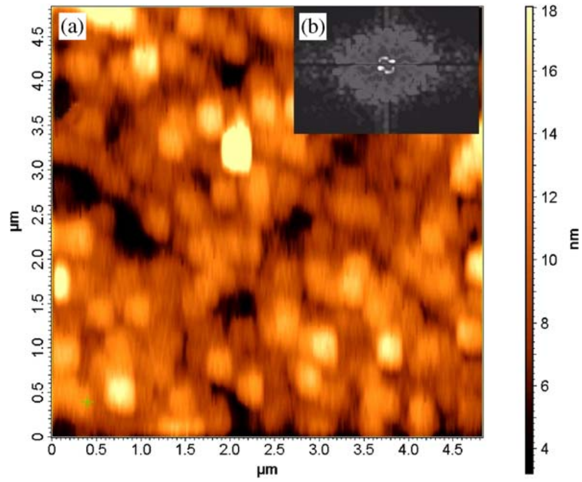

Fig. 3. (a) AFM's surface topography and (b) its FFT spectrum of single-layer TiO_2 films deposited on BK7 glass (amorphous) substrate. The average roughness for this layer is 2.45 nm.

material property and its nucleation process during a particular thin film deposition technique. Whereas, the substrate material decides the nature, symmetry, abundances, and quality of the ordered structures in the thin films/multilayers grown on it. In the stress-driven self-organization process, few earlier experiments indicated that the shapes and sizes of the granular crystallites have a strong layer dependency [24]. Similar effects have been distinctly noticed in our experiments, which are discussed in the following paragraphs.

In order to obtain morphological information on Gd_2O_3 film, we have prepared several samples of single-layer Gd_2O_3, as well as samples in which Gd_2O_3 remained as the surface layer. The topographic measurement demonstrates a random polycrystalline grain structure for this film with a mean roughness value of 3.02 nm. The 2D FFT analysis supports the topographic measurement indicating random polycrystalline structure of the film. The polycrystallinity of the film is reflected as several central bright spots in FFT spectrum that are distributed randomly.

Similarly, in order to observe the topographic nature of TiO_2/SiO_2, several samples of single and multilayers were also prepared. In Figs. 3(a) and (b) the measurement results of a single-layer TiO_2 on BK7 (glass) is presented in order to depict the growth feature of the film as an independent layer. The morphology and 2D FFT analysis results of the film grown on this amorphous substrate has also shown some amount of ordering in the polycrystalline structure.

In order to observe the effect of reactive oxygen partial pressure on the film grain structure, we have prepared two Gd_2O_3/SiO_2 bilayer samples where the surface SiO_2 layer was deposited with and without additional oxygen during the deposition process. The topographic result of the two-layer sample deposited with oxygen (i.e, stoichiometric) is depicted in Fig. 4(a). The SiO_2 grains are highly ordered and sizes are larger compared to the samples depicted in Figs. 1 and 2. The FFT analysis (Fig. 4(b)) of the grain structure shows the presence of both isotropic (diffused ring) and anisotropic (diffraction peaks) components in the structural ordering. The position of the diffraction peaks indicates a different angular symmetry with respect to the earlier grain structure arrangements.

The sample prepared without additional oxygen in SiO_2-layer has been noticed to have ordered grain structure with reduced density. Besides, the grain shape and sizes are observed to be not so regular. This aspect is also highlighted in 2D-FFT

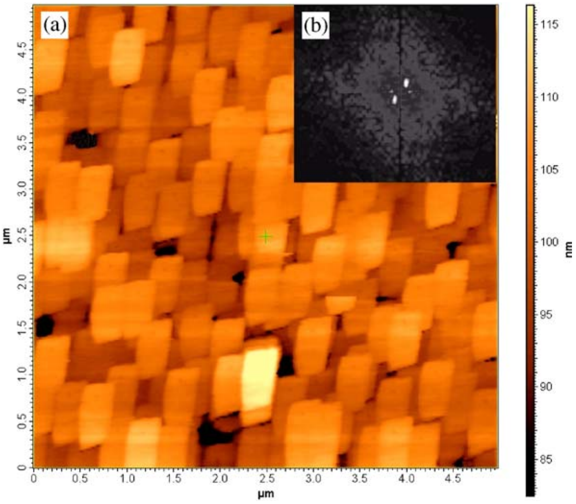

Fig. 4. (a) AFM's surface topographic image of SiO_2 surface self-organized layer (mean roughness is 4.95 nm) and (b) its FFT spectrum in a two-layer (HL) Gd_2O_3(H)/SiO_2(L) multilayer deposited on Si (1 1 1) substrate. H and L correspond to the quarterwave optical thickness of the layers at 248 nm (KrF laser) wavelength. In this case, the top SiO_2 layer has been deposited with optimum oxygen pressure during the growth process.

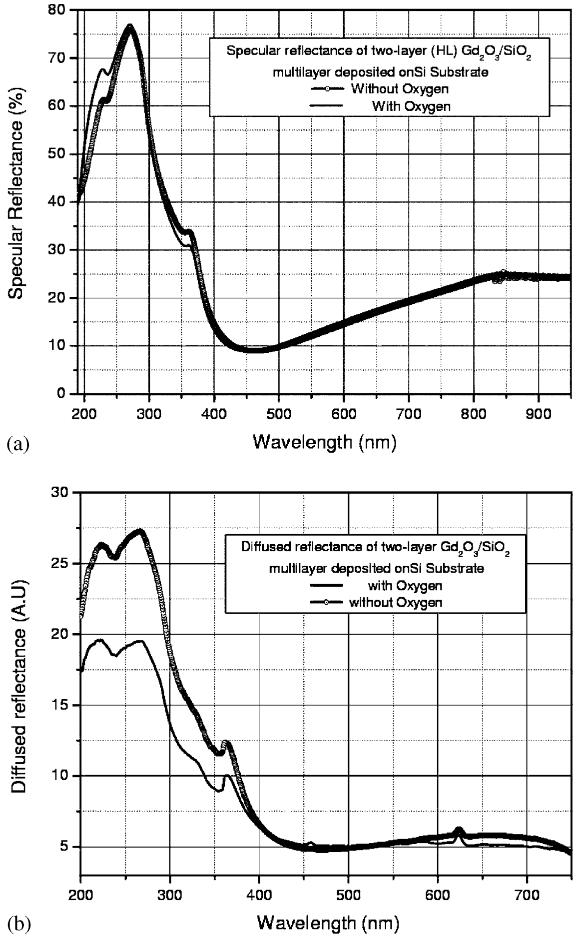

Fig. 5. (a) Specular and (b) diffused reflectance of two-layer (HL) Gd_2O_3(H)/SiO_2(L) multilayer deposited on Si (1 1 1) substrate. Here, the plots with the solid and dotted lines correspond to the SiO_2 component layer deposited with and without oxygen, respectively. Optical thicknesses of the layers are same as in Fig. 4.

spectrum, which has shown both the ring structure, as well as the diffraction peaks. This observation clearly highlights the effect of oxygen partial pressure in the nucleation and surface grain ordering/self-organization process. The specular and diffused reflectance characteristics of these two bi-layer structures are depicted in Figs. 5(a) and (b), respectively. The results of this photometric measurement imply an almost similar effect that is reflected in the topographic measurements relating to grain structure. Both specular and diffused reflection characteristics indicate a better spectral behavior of the coating deposited under oxygen pressure. Superior spectral reflectance for this coating implies a better refractive index, as well as film density under stoichiometric conditions. Higher diffuse spectral reflectance values for the film deposited without oxygen pressure clearly demonstrate the effect of poorly ordered grain structure on the performance factor of optical coating [25].

Fig. 6 depicts the measurement results of a three-layer "LHL" stack, where L and H are SiO_2 and Gd_2O_3, respectively. Here the polycrystalline Gd_2O_3 layer is buried under the self-organized ordered SiO_2 films. The topographic measurement (Fig. 6(a)) shows the presence of highly ordered rectangular grains of homogeneous shape and sizes larger than the single-layer cases. The mean roughness value in the trilayer topography also depicted a much higher value (7.99 nm) than the single layer (4.90 nm). The strain mediation of a polycrystalline layer appears to have a additive effect on the morphology and sizes of SiO_2 grain structures, i.e., almost a two-fold increase in the roughness values (4.9–8 nm) with respect to single layer. Such a drastic change in the topography is due to the change of shape and sizes in self-organized grain structures and may be attributed to a strain field of buried polycrystalline Gd_2O_3 layers. This result resembles the experimental studies carried out by Tersoff et al. [26] in which the grain sizes and shapes are the strong functions of the layer numbers. The FFT result of this measurement is depicted in Fig. 6(b). The analysis results show the presence of both isotropic (diffuse ring) as well as anisotropic components (diffraction peaks) in the grain structure ordering. The orientation of the peaks is very similar to the single-layer SiO_2 on Si(1 1 1) substrate. The topographic measurements have also been carried out on a three-layer system "HLH", which depicted a better value in mean roughness (1.43 nm), which is almost half the single-layer value (3.02 nm). The FFT analysis of this topographic measurement also revealed the polycrystallinity nature of the film along with a better isotropic component for this three-layer design compared to the single-layer

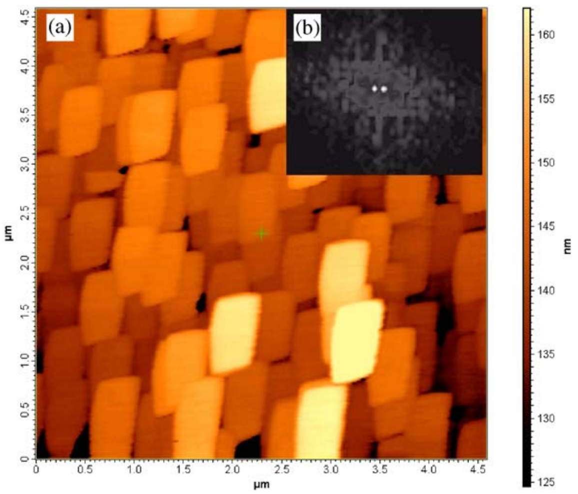

Fig. 6. (a) Surface topography of SiO_2 surface layer (mean roughness is 7.99 nm) and (b) its FFT spectrum in a three-layer (LHL) Gd_2O_3 (H)/SiO_2 (L) multilayer deposited on Si (1 1 1) substrate. Optical thicknesses of the layers are same as in Fig. 4.

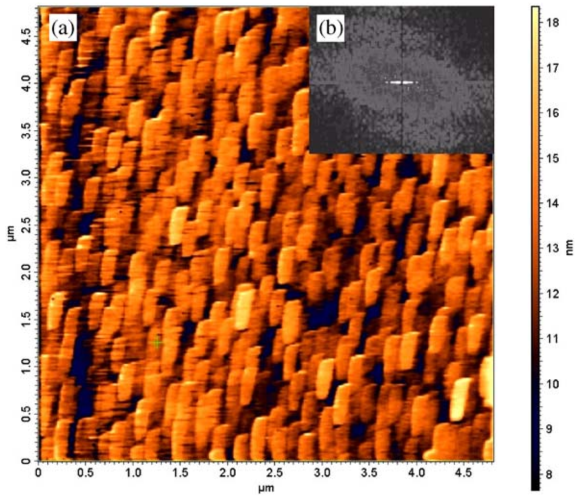

Fig. 7. (a) Surface topography of SiO_2 surface layer (mean roughness is 1.76 nm) and (b) its FFT spectrum in a four-layer (HLHL) Gd_2O_3 (H)/SiO_2 (L) multilayer deposited on Si (1 1 1) substrate. Such a four-layer design has shown better self-organized grain structure as well as morphology. Optical thicknesses of the layers are same as in Fig. 4.

Table 1
The statistics on the roughness values for different single and multilayer films deposited on Si (1 1 1) substrate signifying strain mediation effects

S. No.	Layer type	Roughness values		Remarks
		Mean (nm)	RMS (nm)	
1	[Si]–SiO_2	4.9	6.21	Single-layer "SiO_2"
2	[Si]–SiO_2–Gd_2O_3–SiO_2	7.99	11.25	Gd_2O_3 layer buried in between SiO_2 layers
3	[Si]–Gd_2O_3	3.02	3.87	Single-layer "Gd_2O_3"
4	[Si]–Gd_2O_3–SiO_2–Gd_2O_3	1.43	1.7	SiO_2 layer buried in between Gd_2O_3 layers

one. These experiments indicate that a buried ordered structure layer can improve the quality (morphology) of a polycrystalline or amorphous layer, which can be related, to better and effective strain mediation of a self-organized grain structured film. The statistics of the roughness mesurements on single and tri-layer structures are presented in Table 1. The data values indicate that a superior RMS roughness can be achieved with the buried SiO_2 layer in between Gd_2O_3 layers.

The next result in Fig. 7 depicts the topographic as well as FFT results of a four-layer ‘‘HLHL’’ structure deposited on silicon substrate. Such a design has possibly carried the combined strain-mediation effects of both ordered SiO_2 and polycrystalline Gd_2O_3 layers. The grains appear to be more regular and better ordered but mean grain sizes become smaller than the ‘‘LHL’’ multilayers. The improved ordering is also reflected in the FFT results in Fig. 7(b). It can be noticed from this figure that both the isotropic and anisotropic components in the grain ordering are more prominent. The statistics on the grain-structure geometries for these single- to four-layer devices have been presented in Table 2. The results of this four-layer system clearly point out the scope of achieving superior morphology by adopting appropriate combination of self-organized and polycrystalline films in multilayers.

Besides using Gd_2O_3/SiO_2 combinations, we wanted to verify the self-organized ordered grain-structure symmetry in SiO_2 by using a multilayer combination with different material other than Gd_2O_3 and on amorphous substrate. We prepared a four-layer sample of ‘‘HLHL’’ device on a glass substrate using TiO_2/SiO_2 combinations where H, L corresponds to TiO_2 and SiO_2, respectively. The topographic measurement (Fig. 8(a)) showed a similar high-quality ordering in the surface grain structure of the top SiO_2 film. The FFT analysis (Fig. 8(b)) of this measurement showed the contributions of isotropic and anisotropic components with an overall distinct four-fold symmetry in the grain structure. The results of these experiments indicated that the initiation of self-organized surface grain structures in SiO_2 films is primarily owed to the nucleation and growth process under the present experimental process parameters. It might be the reason of observing such organized structures and morphology when

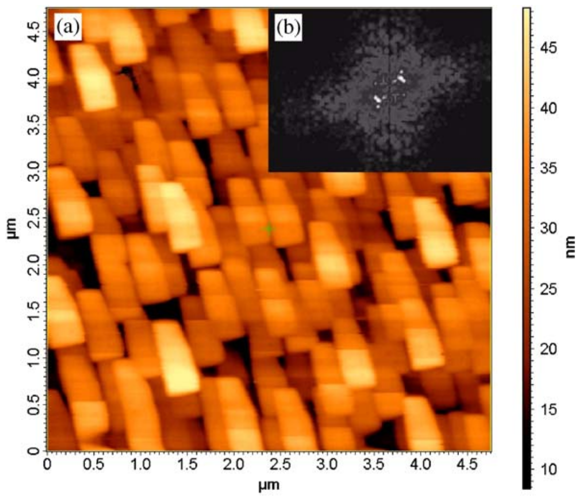

Fig. 8. (a) Surface topography of SiO_2 self-organized surface layer (mean roughness is 6.46 nm) and (b) its FFT spectrum in a four-layer (HLHL) TiO_2 (H)/SiO_2 (L) multilayer deposited on BK7 glass (amorphous) substrate.

Table 2
The statistics on the mean grain sizes for different single and multilayer films deposited on Si (1 1 1) substrate

S. No.	Layer type	Grain size information (mean values)		
		X (nm)	Y (nm)	Grain area $\times 10^4$ nm^2
1	[Si]–SiO_2	320	226	7.2320
2	[Si]–Gd_2O_3–SiO_2	390	604	23.5560
3	[Si]–SiO_2–Gd_2O_3–SiO_2	557	775	43.1675
4	[Si]–Gd_2O_3–SiO_2–Gd_2O_3–SiO_2	150	414	6.2100

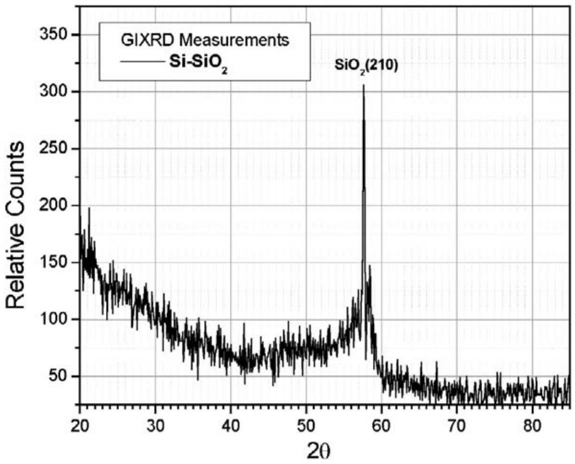

Fig. 9. GIXRD measurement of a single-layer SiO_2 film on silicon substrate depicting a sharp and single SiO_2 (2 1 0) hexagonal peak. Angle of incidence was chosen to be 0.4° for the measurement.

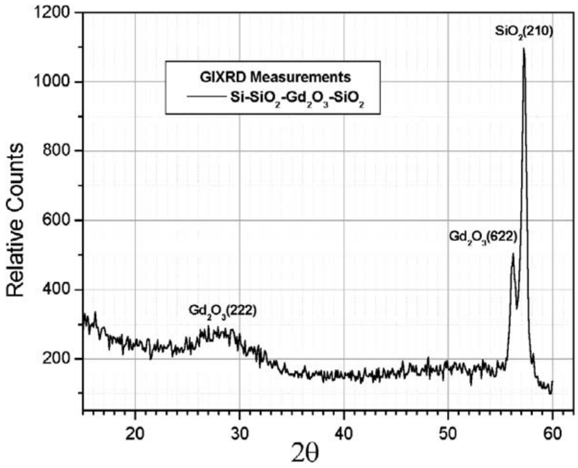

Fig. 10. GIXRD measurement of a three-layer Si–SiO_2–Gd_2O_3–SiO_2 sample depicting the presence of ordered SiO_2 as well as polycrystalline Gd_2O_3 (cubic) peaks. The SiO_2 (2 1 0) peak signal has been substantially increased due to the presence of two ordered structure SiO_2 layers in the tri-layer structure.

the films were grown on amorphous glass substrates and also along with TiO_2 or Gd_2O_3 as associated layers.

4.2. Results of the GIXRD measurements

The GIXRD measurement results of two samples are discussed in the present paper. The two sample geometries selected for this analysis are (i) Si–SiO_2 and (ii) Si–SiO_2–Gd_2O_3–SiO_2, respectively. For GIXRD measurements, the glancing angle of incidence have been chosen to be 0.4° and the diffraction angle (2Θ) are scanned from 10° to 85°. The result of the single-layer Si–SiO_2 sample depicted in Fig. 9 has shown only one diffraction peak corresponding to SiO_2 (2 1 0). The single and sharp peak distinctly supports the presence of highly ordered grain structure. The next sample, which carries the layer geometry of Si–SiO_2–Gd_2O_3–SiO_2 has a buried polycrystalline gadolinia layer. The GIXRD measurement clearly reveals the presence of the both ordered-structured SiO_2 and the polycrystalline gadolinia (cubic) component layers. In this result (Fig. 10), besides the SiO_2 (2 1 0) peak at 57.2°, the additional two peaks at 56.2° and 28.5° belong to the cubic Gd_2O_3 and correspond to the ($h\,k\,l$) value of (6 2 2) and (2 2 2), respectively. It is interesting to note that the peak amplitude of SiO_2 (2 1 0) has been substantially enhanced due to the presence of two SiO_2 ordered structured layers in the sample geometry. This result also indicates that the buried SiO_2 layer has also carried the ordered grain structures like the surface one.

5. Conclusions

SiO_2 films deposited by reactive electron beam deposition process have depicted a novel self-organized surface grain structures both in single and multilayer geometry. Probing such structures especially under varied multilayer geometry is a challenging as well as an interesting task. Regular and ordered microcrystalline surface grain structures were verified by multimode scanning probe microscopy, 2D power spectral FFT analyses and GIXRD measurements. The highly reproducible ordered grain-structure symmetries under various multilayer geometries can be primarily attributed to the strain mediation as well as stress-relief mechanisms in their growth process. Various experimental observations through topographic and spectral measurements indicated that formation of such ordered structure in nonepitaxial thin films exclusively depends upon the combined effect of the individual material parameters and its process-dependent nucleation properties. Stoichiometry has played a very important role in the self-organization processes. The nature of the substrate and the associated layers in multilayer geometries has shown to affect shape, size, angular symmetry and abundance of the ordered structures. The strain mediations of ordered structured layer as buried layers depicted superior surface morphology of polycrystalline layers. More deep insight into the topic will generate a better understanding on the achievement as well as control over the grain size ordering and multilayer topography, which, sometimes, are the essential factors in developing scatter-free optical coatings and devices.

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