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Nano-structure and optical properties (plasmonic) of graded helical square tower-like (terraced) Mn sculptured thin films

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Graded helical square tower-like terraced sculptured Mn thin films (GHSTTS) are produced in three stages with different number of arms using oblique angle deposition together with rotation of substrate holder about its surface normal, plus a shadowing block fixed at the centre of the substrate holder. The structural characterization of the produced samples was obtained using field emission scanning electron microscope (FESEM) and atomic force microscope (AFM). Results showed a structural gradient with distance from the edge of the shadowing block, which in turn is responsible for the decrease in the volume of void fraction and increase of grain size. Plasmon absorption peaks observed in the optical analysis of these nano-structures showed that their wavelength region and intensity depend on the polarization and the incident angle of light, as well as the distance from the edge of the shadowing block. According to our model and discrete dipole approximation (DDA) calculations, when the number of parallel nano-rods of different lengths and radii are increased the peak in the spectrum shifts to shorter wavelengths (blue shift). Also in the diameters of the nano-rods increases (a situation that occurs with increasing film thickness) the results is again a blue shift in the spectrum. The presence of defects in these sculptured structures caused by the shadowing effect is predicted by the theoretical DDA investigation of their optical spectra. Good agreement is obtained between our theoretical results and the experimental observations in this work.

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1. Introduction

The geometrical shape and size of nano-structures have direct influence on their different properties. This has interested many researchers to investigate the properties of these structures in different areas of science such as optics, electronics and biophysics. Nanostructures of noble metals (i.e., Au, Ag and Cu) have attracted researchers’ attention because of their localized surface Plasmon resonance (LSPR) property. When the electrons in nano-structured noble metals are excited by incident light, optical resonances occur because of the collective oscillation of conduction electrons. This collective excitation is known as localized surface plasmon [1]. At these incident light wavelengths where the surface Plasmon resonance occurs the surface electric field increases, which is the basis of surface enhanced Raman spectroscopy [2,3]. LSPR depends on the composition [4–6], surrounding environment [6] and geometry of the metallic nano-structure [7–9].

This phenomenon because of its numerous applications has been subject of many investigations, [e.g.10–17] most of which are concentrated on noble metals (gold and silver). The reason for choosing noble metals is that they show many oscillations and a varying refractive index [18], which in turn are responsible for sharp changes in the optical spectra of these materials as thin films, leading to pronounced Plasmon peaks in their extinction spectra. However, the geometrical structure of nano-particles from which the thin film is formed also has a strong influence on the plasmonic properties although they may be made of the same material [7,17,19]. Therefore, we may be able to fabricate nano-structures with as many as possible sharp corners or protruding tips acting as hot spots for increasing the electric field at these points.

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By changing the polarization of the incident light we may adjust and control the plasmonic peaks. Therefore, by fabricating sculptured thin films with different geometrical shapes we may investigate the influence of shape on the plasmonic properties, irrespective of the material itself. Many researchers have previously worked and reported on the shape and geometry of single nano-particles or the effect of the surrounding medium on the behaviour of a single nano-particle [20–22]. However, to the best of our knowledge, we may only refer to two works which extended this kind of study to an assembly of sculptured nano-particles which form the structure of the thin film rather than a single nano-particle [19, 23].

In recent years the oblique angle deposition technique (OAD) has provided the facility for production of different nano-structures with varying anisotropy [24]. OAD (vapour incident angle less than 85°) and glancing angle deposition (vapour incident angle greater than 85°) together with rotation of the substrate holder about its surface normal are used for fabrication of 3D nano-structures in different fields of study [25, 26].

Brett and Krause [27] by using the OAD technique in conjunction with rotation of the substrate holder about its surface normal while fixing a shadowing object/block at the centre of the substrate holder, produced helical nano-structures called graded chiral nano-sculptured thin films. Savaloni et al. [28] used the same kind of setup to produce graded chiral zig-zag silver sculptured thin films. The slope of these graded structures decreases on increasing the distance from the shadowing block, hence their nano-structure and their different properties varies with distance from the shadowing block. The optical behaviour of these structures is studied theoretically by Babaei et al. [29].

In our earlier work [23] we used manganese, rather than a noble metal and produced different shaped nano-particles (i.e., helical rectangles and helical pentagon) and investigated their plasmonic properties by introducing a model for the sculptured structures made of nano-rods, and considered all of the possible states (positions) that one nano-rod may have with relation to its neighbouring nano-rods in the helical rectangles and helical pentagon (see Fig. 7 in Ref. [23]). In the extinction spectra of simple Mn nano-rods, pronounced plasmon peaks were not observed [30] because the refractive index of Mn, unlike noble metals, does not show much variation with wavelength [18]. As mentioned in Ref. [23] the use of Mn should not restrict our findings to this material as the geometry of the structure is the main feature that influences the results.

In the present work, the geometry of the Mn nano-particles within the structure of our thin films is changed to graded helical square tower-like terraced shape (GHSTTS), which is totally different from that discussed earlier and so the model for calculation of extinction spectra presented in our earlier work [23] is extended here to cover the situations created by this geometry.

The graded helical square tower-like terraced shaped sculptured thin films (GHSTTS) were produced by oblique angle deposition together with the rotation of the substrate sequentially by 90°, while a shadowing block was fixed at the centre of the substrate holder having substrates mounted on it at different distances from the edge of the shadowing block. In addition the length of the arms of the second and third pitches of the GHSTTS were reduced by a third and two thirds relative to the length of the arms of the first pitch, hence terraced structures as well as slanted shapes in which the tilt angle changes with distance from the edge of the shadowing block were formed. Hence, their structure provides numerous hot spots which in turn can be responsible for the plasmonic resonances.

We used the discrete dipole approximation (DDA) method to simulate and calculate the scattering cross-section of Mn nano-rods in the form of combinations of different number of nano-rods with different lengths and diameters as well as different tilt (growth) angles.

The structural characteristics of these nano-sculptured thin films are analysed, while their optical properties for different polarizations and different incidence light angles and azimuthal angle as a function of distance from the edge of the shadowing block are also studied. The structures of GHSTTS produced in this work are periodical in two normal and parallel directions with respect to the substrate surface, are anisotropic and as mentioned above are grown with graded growth angle with distance from the edge of the shadowing block. Hence, the surface plasmon resonance peaks can be adjusted by distances from the edge of the shadowing block and using different incidence angles of polarized light.

Fig. 1. Simulation figures of helical square structure with the corresponding view directions. L and r are the length and the radius of each nano-rod, respectively. αi is the growth angle of helical columns. The definition of the incident polarization directions as well as decomposition of the p-polarized field by changing the incident light direction is shown in (c).
2. Theory

2.1. The discrete dipole approximation (DDA)

Detailed description of the DDA is given in many published papers [e.g., 31–33], and calculations for different nanoparticles are also given in many literatures. In particular, with regards to helical structures having different lateral cross-sections (i.e., square, rectangle, and pentagon) and whose building blocks are nano-rods, a detailed discussion and model is given in Ref. [23]. In a model they have considered all of the possible states (positions) that a nano-rod may have with its neighbouring nano-rods (see Fig. 7 in Ref. [23] and discussion therein). However, that work did not consider multiple (more than two) parallel nano-rods of different sizes (i.e., lengths.

![Image](image_url)
and radii), the distances between them and the tilt angle of the helical structure (situations that occur for the samples positioned close to the shadowing block in this work), which all apply to the samples made in this work as graded helical square tower-like terraced shape (GHSTTS) sculptured structures. Therefore, we should examine theoretically these different states of our samples before being able to analyse our optical results. In the following paragraphs we report on the obtained DDA results for these conditions, while a brief discussion of the DDA theory is also given.

The DDA deals with nano-particles of arbitrary shape and with size of the order of, or smaller than, the wavelength of the incident light [31–33].

An assembly of discrete dipoles fills the body of the nanoparticle. The interaction of the electric field of the incident light with these dipoles as well as the interaction of these dipoles with each other is taken into calculation in the DDA. Hence, an external electric field produces a local electric field at the position of each point dipole. The remaining dipoles also produce an electric field at this point. Therefore, each dipole effectively experiences an electrical dipole moment which is due to the local electric field of the above mentioned process. Calculation of this dipole moment enables us to obtain responses to the scattering problems [31–33].

The mean separation distance between dipoles, \( d \) in the assembly can be obtained as:
\[
d = \left( \frac{V}{N} \right)^{1/3},
\]
where \( N \) is the number of point dipoles and \( V \) is the volume of the object (nanoparticle). The dipole is considered as a sphere with a refractive index, \( m \). All dipoles become polarized by the electric field of the incident light. The method of calculation for these polarizations is given in Refs. [31–33]. The total scattering cross section as a measure of the extinction spectrum can be obtained using the optical theory, giving:
\[
C_{\text{ext}} = \frac{4\pi k}{|E_0|^2} \sum_{j=1}^{N} \text{Im} \left( E_{\text{inc},j} \cdot P_j \right),
\]
where \( E_0 \) is the amplitude of the incident electric field which is considered as unity in the calculations, \( k = \frac{2\pi \lambda}{\lambda} \) is the wave number, \( E_{\text{inc},j} \) is the electric field of the incident light at the location of the jth dipole, and \( P_j \) is the polarization of the jth dipole as a result of the electric field of the incident light at the jth dipole location plus the electric field of all other dipoles except jth dipole induced at the jth dipole location. The number of dipoles substituted in
the body of the object (nanoparticle) determines the accuracy of the method. The convergence limit for this method is given as \( m k d < 1 \). As suggested by Siabi and Savaloni [23], considering that the sculptured thin film itself is an assembly of nanoparticles and since in the DDA method the interaction between these nanoparticles decreases with increasing distance between them, we may consider the obtained spectrum from a thin film as an overlap of spectra of nanoparticles constituting the thin film.

In Fig. 1 the simulation of GHSTTS with its corresponding view directions is given. \( L \) and \( r \) are the length and the radius of each nano-rod, respectively.

Our proposed model for our present samples possessing differing numbers of parallel nano-rods with different dimensions, spacings, and tilt angle of the helical structure, together with the incident light direction is shown in Fig. 2(i–iii). This model may be considered as an extension to that of Siabi and Savaloni [23]. As pointed out by Siabi and Savaloni [23], in all calculations the actual orientation of the electric field of the incident light with respect to the geometrical structures must be considered, rather than simply the polarization of the incident light (i.e., s- or p-polarization). The distance between dipoles is taken to be \( d = 1 \) nm and all optical constants for Mn were obtained from [18].

In Fig. 2(i–iii), the calculated scattering cross-sections for two and three parallel nano-rods with different dimensions and spacings are also given. In Fig. 2(i–a), the electric field of the incident light is considered to be in the same direction as that of the nano-rods \((\chi = 34^\circ; \theta = 34^\circ)\).
Comparison of the calculated scattering cross-section spectra (Fig. 2(i-b)) of two parallel nano-rods noted as case 1 in Table 1 (with a peak formed at 440 nm) with two other parallel nano-rods noted as case 2 (with a peak formed at 342 nm) in Table 1 shows that the peak of the spectrum of the latter case is shifted to shorter wavelengths (blue shift). Hence when the thickness of the shorter nano-rod increases a blue shift is observed.

In Fig. 2(i-c) and (i-d) the near-field electric contours distributions for the two parallel nano-rods (case 1 in Table 1) and (case 2 in Table 1) where the electric field of the incident light is along the nano-rods axes are given, respectively. In Fig. 2(i-c) we observe two dark areas in the space between the two nano-rods, indicative of quadrupole oscillations. Similar results can be seen in Fig. 2(i-d) with somewhat lower intensities and slightly closer dark areas.

Comparison of the calculated scattering cross-section spectra (Fig. 2(ii)) obtained for three parallel nano-rods noted as case 1 in Table 2 with three other parallel nano-rods noted as case 2 in Table 2 shows that the peak of the spectrum of the latter case is again shifted to shorter wavelengths (blue shift). Therefore, it can be deduced that the addition of more nano-rods, shorter in length and thicker in diameter, leads to higher degree of blue shift. It may be speculated that by increasing the diameter of the nano-rods the peak of the scattering cross-section undergoes a blue shift. Because the position of the peak in these spectra cannot be clearly distinguished, one cannot calculate the near-field contour distribution.

Since the structures in this investigation have different tilt angles when formed at different distances from the edge of the shadowing block it was also decided to investigate the influence of this parameter on the results. In Fig. 2(iii-a) the calculated scattering cross-sections for three parallel nano-rods of different lengths and different diameters (Table 2), when \( \chi = 0^\circ, 20^\circ, 34^\circ, 55^\circ, 70^\circ \), are given. For the first three angles (i.e., \( 20^\circ, 34^\circ \) and \( 55^\circ \)) very little change can be observed. However when the angle is increased to \( 70^\circ \) a peak appears at \( \sim 330 \) nm. Hence, it may be suggested that when the structure (specifically the arms of the structure) is tilted to very high degrees the peak of the spectrum undergoes a red-shift. The near-field electric contours distribution for the peak observed in case of \( \chi = 0^\circ = 70^\circ \) by assuming that the peak position is at \( 330 \) nm is given in Fig. 2(iii-b). The maximum intensity of the electric fields is at the two ends of the nano-rods. Therefore, between the top edge of nano-rod 1 and the bottom edge of nano-rod 2 there is a strong electric field interaction and a quadrupole oscillation occurs (two dark spots can be distinguished between these two nano-rods).
Hence, is with the growth/rise of nano-rods, the parallel edge spacings become shorter and the bottom edges of nano-rods 3 which is a short and fat nano-rod. When the angle is reduced the peaks shifts towards shorter wavelength and cannot be observed in the spectral range shown in Fig. 2(iii-a). Hence, the near-field electric contours distribution for these spectra may not be presented. Therefore, it may be concluded that between two parallel nano-rods with different lengths (e.g., 36 and 72 nm) a quadrupole oscillation occurred (two dark spots between these two nan-rods forms), but when the tilt angle (χ) of these nano-rods increases to 70° the space between these nano-rods reduces and only a dipole oscillation can be observed.

The extinction spectra obtained from three parallel nano-rods with different lengths and radii when they are tilted with respect to the direction of the incident electric field are given in Fig. 2(iii-c). The growth/rise angle of the nano-rods as mentioned throughout this work are fixed at χ = 34°. The incident light angle is changed so that the angle between the incident electric field and the nano-rods is changed (ζ) (e.g., when the incident light angle is at θ = 90° this angle becomes ζ = 90° − 34° = 56°), hence, when the angle between the incident light and the axis of the nano-rod increases, the extinction (scattering cross-section) decreases and the peak undergoes a blue shift.

Again if we compare all the results for two and three parallel nano-rods in Fig. 2 it can be deduced that the spectra of three parallel nano-rods with different lengths and radii and different spacings (i.e., the actual situation of our samples in this work) show higher degree of blue shift relative to the two parallel nano-rods of different lengths and radii.

The effect of varying the incident light electric field orientation (or tilt angle of nano-rods relative to the incident light) on the scattering cross-section of three nano-rods similar to the case 2 in Table 2 is shown in Fig. 2(iii-c). It can be seen that as the angle between the electric field of the incident light and the axis of the nano-rods (ζ) increases the scattering cross-section decreases and the peak of the spectrum shifts to shorter wavelengths (blue shift). The conclusion from the results for three parallel nano-rods with different length, radii and spacing are the same as that Siabi and Savaloni [23] reached for two parallel similar nano-rods: when

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**Table 2**

Details of structural parameters for three parallel nano-rods.

<table>
<thead>
<tr>
<th>Parallel Nano-rods</th>
<th>Nano-rod 1</th>
<th>Nano-rod 2</th>
<th>Nano-rod 3</th>
<th>D (nm)</th>
<th>D' (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>L = 110 nm; r = 6 nm</td>
<td>L = 72 nm; r = 6 nm</td>
<td>L = 36 nm; r = 6 nm</td>
<td>82</td>
<td>53</td>
</tr>
<tr>
<td>Case 2</td>
<td>L = 110 nm; r = 6 nm</td>
<td>L = 72 nm; r = 8 nm</td>
<td>L = 36 nm; r = 10 nm</td>
<td>82</td>
<td>53</td>
</tr>
</tbody>
</table>

D is the distance between nano-rod 1 and nano-rod 2, D’ is the distance between nano-rod 2 and nano-rod 3.
Hence, and motors block.

ligible surface movement (electric slide) (GHSTTS) production of H9251 and behaviour of Edwards × field = H9258. Experimental 10 nano-rods of field = H9264. Substrates of substrate for deposition was H9272. The 7 mbar normal (GHSTTS) was observed. Nano-rods of substrate with 0.01 ◦ accuracy between the substrate deposition rate was measured for a quartz crystal deposition rate controller (Sigma Instruments, SQM-160, USA) positioned close to the substrate holder and at almost the same azimuthal.

3. Experimental details

The graded helical square tower-like (terraced) sculptured (GHSTTS) Mn thin films were deposited on glass (microscope slide) substrates by electron beam evaporation from a graphite crucible at room temperature. The purity of Mn was 99.99%. An Edwards (Edwards E19 A3) coating plant with a base pressure of 2 × 10⁻² mbar was used. The deposition angle was fixed at 80◦ and the substrate was rotated clock-wise for 90° azimuthally for production of each arm of the GHSTTS. Fig. 3(a and b) show the schematic of the deposition set up with substrate position and rotation for sculptured helical thin film growth as well as growth behaviour of GHSTTS structures with distance from the shadowing block. The substrate holder system is controlled by two stepping motors which can rotate the substrate holder by the two angles, α and ϕ, with 0.01 ◦/step accuracy and with controlled speed. The movement of the stepper motor for rotation of substrate about its surface normal (ϕ) and its speed of revolution as well as facility for dividing each revolution to different sectors are controlled through interface to a computer in which the related software (in the LABVIEW format) is written and installed. All these are domestic made. The substrate holder was a stainless steel disc of 12 cm diameter. At the centre of this disc a cylindrical block (2 cm in diameter and 2 cm in height) was fixed as a shadowing block. The substrates were fixed at 1, 3 and 5 cm distance from this shadowing block along four mutually normal radii of the substrate holder disc. Hence, in each run four sets of three samples were produced for use in different analyses and reproducibility check of the samples.

Three different GHSTTS Mn thin films consisting of different number of arms, namely eight, nine and ten arms were produced. Each arm of the first pitch was deposited for 110 nm while each arm of the second and third pitches was 72 nm and 36 nm, respectively.

In oblique angle deposition the angle between the axis of columns and normal to the substrate surface “β”, when the deposition angle “α” is larger than 60◦ may be obtained from the Tait [34] equation as:

\[ \beta = \alpha - \sin^{-1}\left(\frac{1 - \cos \alpha}{2}\right) \]  

Accordingly, β = 55.5◦ was obtained and the film thickness (normal to the substrate surface) for each arm of the first pitch was 62.30 nm and in the second pitch and third pitch it was 40.78 nm and 20.39 nm, respectively. The deposition rate was fixed at 1.0 Å s⁻¹. The deposition rate was measured by a quartz crystal deposition rate controller.
Table 3
Details of graded helical sculptured Mn thin film samples and their different parameters.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Distance from the shadowing block (cm)</th>
<th>Film thickness (nm)</th>
<th>Helical column tilt angle ($\alpha_t$) (°)</th>
<th>D$_{AFM}$ (nm)</th>
<th>R$_{ave}$ (nm)</th>
<th>R$_{rms}$ (nm)</th>
<th>VF$_{surf}$ (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A1</td>
<td>1</td>
<td>465.5</td>
<td>14</td>
<td>122.0</td>
<td>1.75</td>
<td>2.24</td>
<td>13.32</td>
</tr>
<tr>
<td>A3</td>
<td>3</td>
<td>504.1</td>
<td>8</td>
<td>112.0</td>
<td>1.33</td>
<td>1.68</td>
<td>13.19</td>
</tr>
<tr>
<td>A5</td>
<td>5</td>
<td>550.8</td>
<td>4</td>
<td>96.0</td>
<td>1.20</td>
<td>1.50</td>
<td>9.60</td>
</tr>
<tr>
<td>B1</td>
<td>1</td>
<td>436.7</td>
<td>12</td>
<td>120.7</td>
<td>2.33</td>
<td>2.87</td>
<td>19.04</td>
</tr>
<tr>
<td>B3</td>
<td>3</td>
<td>450.0</td>
<td>8</td>
<td>109.6</td>
<td>2.20</td>
<td>2.86</td>
<td>16.44</td>
</tr>
<tr>
<td>B5</td>
<td>5</td>
<td>544.8</td>
<td>4</td>
<td>81.0</td>
<td>1.96</td>
<td>2.58</td>
<td>12.87</td>
</tr>
<tr>
<td>C1</td>
<td>1</td>
<td>417.4</td>
<td>12</td>
<td>132.3</td>
<td>2.60</td>
<td>3.42</td>
<td>22.92</td>
</tr>
<tr>
<td>C3</td>
<td>3</td>
<td>441.0</td>
<td>5</td>
<td>128.9</td>
<td>2.57</td>
<td>3.28</td>
<td>29.32</td>
</tr>
<tr>
<td>C5</td>
<td>5</td>
<td>525.0</td>
<td>3</td>
<td>91.9</td>
<td>2.03</td>
<td>2.60</td>
<td>19.55</td>
</tr>
</tbody>
</table>

D$_{AFM}$: Grain size obtained from AFM analysis. R$_{ave}$ and R$_{rms}$: Average and root mean square surface roughnesses, respectively. VF$_{surf}$: Surface void fraction.

angle as that of the substrate. This was calibrated after obtaining the film thickness using a field emission electron microscope (FESEM) and dividing by the deposition time. The distance between the evaporation source and the substrate was 30 cm. In this arrangement the vapour source (6 mm in diameter) behaves like a point source with a cosine distribution and because of the 30 cm distance between the vapour source and the substrate it is expected that the vapour has straight trajectories (i.e., no appreciable scattering due to the large mean free path ($\sim$ 103–104 cm [35]) occurs).

Prior to deposition, all glass substrates were ultrasonically cleaned in heated acetone then ethanol. The surface roughness of the substrates was measured by a Talysurf profilometer and AFM

![Fig. 6](image-url)

(i) Optical spectra obtained at $\theta = 10^\circ$ incident light angle using both s- and p-polarized lights and different azimuthal angles ($\varphi$) for the graded helical square tower-like (terraced) Mn sculptured thin films produced at different distances (D) from the edge of the shadowing block with 10 arms. (ii) Optical spectra obtained at $\theta = 60^\circ$ incident light angle using both s- and p-polarized lights and different azimuthal angles ($\varphi$) for the graded helical square tower-like (terraced) Mn sculptured thin films produced at different distances (D) from the edge of the shadowing block with 10 arms.
and the rms (route mean square) substrate roughness (Rq) obtained using these methods was 0.3 nm and 0.9 nm, respectively.

The deposition process was repeated a few times and the reproducibility of the results (i.e., optical reflection from similar films produced) was confirmed. The film thicknesses and column shapes and sizes were measured by field emission electron microscope (FESEM) (Hitachi S-4100 SEM, Japan). The FESEM samples were coated with a very thin layer of gold to prevent the charging effect.

The surface physical morphology and surface roughness of the samples were obtained by means of AFM (NT-MDT, SOLVER, Nova Tech) analysis with a Si tip of 10 nm in diameter and in non-contact mode. The optical spectra of the samples were obtained using a single beam spectrophotometer (Aquila Instruments, NKD-8000) in the spectral range of (350–1100 nm) using both s and p polarization measurements in steps of 5 nm and at two incidence angles of 10 and 60°.

4. Results and discussions

4.1. Structure and surface morphology of the graded helical nano-sculptured Mn thin films

Fig. 4 (columns I to III) shows the FESEM images of the cross sections of the GHSTTS Mn thin films with eight, nine and ten arms produced at three different distances of 1, 3 and 5 cm from the edge of the shadowing block. The helical structure of the grown nano-columns and the dense bottom layer of the films [36,37] can clearly be distinguished in these images, while the grown arms of the square pitches and their angle as well as the growth angle of the columns (tilt angle) are also seen. In addition one can also see the reduction of the length of the arms from bottom pitch to upper pitches consistent with the deposition procedure (110 nm, 72 nm and 36 nm lengths of arms for the bottom, middle and top pitches, respectively). The tilt angle of the helical columns is measured from the FESEM images and is given in Table 3 for GHSTTSs with eight, nine and ten arms as mentioned above.

In order to clearly identify the produced samples with three different number of arms and that each set contains of three different samples which were positioned at three different distances from the edge of the shadowing block (i.e., 1, 3 and 5 cm) we have assigned the following names/symbols to them:

The set with 10 arms are assigned as “A” group followed by distance from the shadowing block in centimetres: A1, A3 and A5. Similarly for the set with 9 arms we assigned them as B1, B3 and B5 and for the group with 8 arms we named them as C1, C3 and C5. Results of growth/tilt angle of columns and the thickness of the films measured from the FESEM images are given in Table 3 and these are consistent with the calculated results.
The shadowing block is responsible for the variation in the growth of these slanted helical sculptured structures and it can be observed that the tilt angle \( (\alpha_t) \) decreases and the diameter of the helical structures increases with increasing distance from the edge of the shadowing block. A detailed description of the effect of the shadowing block on the deposition process under the set up described in this work is given in our earlier works [28,38].

Fig. 5(i–iii) show the 2D and 3D AFM images (columns I and II) as well as surface void/inclusion fractions (column III), while in Table 3 the results of grain size distribution, percentage of surface void fraction (measured using JMicroVision software), average and root mean square surface roughness for all samples produced in this work obtained from the AFM results are given.

Results in Table 3 for each group of samples (i.e., A1–A5, B1–B5 and C1–C5), show that the surface fraction of voids and the surface roughness of the samples are reduced by increasing the distance from the edge of the shadowing block. This is consistent with the results reported in Refs. [28,38]. In addition, it can also be seen that the surface roughness is reduced by increasing the number of arms for corresponding distances from the edge of the shadowing block. This is due to the fact that by increasing the number of arms, smaller and broader arms are formed on top of the film, hence covering the surface of the film more uniformly.

The other point which is worth mentioning is that the features (grains) are inclined towards the higher vapour flux on the surface of the films produced nearer to the edge of the shadowing block (see 3D AFM images in Fig. 5(i)–(iii)). Hence from these observations it can be deduced that one can design and fabricate various sculptured thin film structures with tuning properties, using the deposition technique mentioned in this work.

4.2. Optical characterization

The schematic for the GHSTTS structure and the definition for the incident light polarization directions as well as decomposition of the \( p \)-polarized field on changing the incident light direction is shown in Fig. 1. As illustrated in Fig. 1 and discussed in Ref. [39], when the incident light interacts with the GHSTTS structure at an angle the \( E_p \) field can be decomposed into two components: a field parallel \( E_{ph} \) to, and a field perpendicular \( E_{pl} \) to the long axis of this
structure. Therefore, the larger $\theta_1$ results in a larger $E_{\text{all}}$ component (Fig. 1). This means that the results from a low incidence angle ($10^\circ$) should contain information about the surface morphology of the film, including anisotropy when the $\varphi$ angle is changed; while high incidence angle ($60^\circ$) light interacts with the Mn column with its maximum electric field. For s-polarization, when the $\varphi$ angle is changed, because of possible surface anisotropy of square, we should obtain some information about the surface morphology of the film too.

Because of possible asymmetry along the helical axis of the square sculptured thin film, particularly for those structures produced at close distance to the shadowing block that grow with an angle to the surface normal (i.e., tilted tower-like (terraced) helical square structures) we may observe a change in the peak position which can be due to the changes of angle between the incident light electric field direction and the effective arm(s) of the structure. The arms of these particular structures, depending on the number of arms, take different orientations with respect to the direction of the incident light components as well as the length(s) of the arm(s). This will be discussed in detail in the following text.

Optical responses of the samples produced in this work to both s- and p-polarizations were measured at two incident light angles of $10^\circ$, and $60^\circ$ in steps of $5$ nm and at different azimuthal angles of $0^\circ$, $45^\circ$, $90^\circ$, $120^\circ$, $135^\circ$, $150^\circ$ and $180^\circ$. The transmission of all samples was close to zero (apart from those that showed small values at long wavelengths, which will be mentioned during the discussion of their results). This is consistent with the film thickness of the samples given in Table 3. With good approximation we may assign (1-R-T) as a good qualitative estimate for the absorption. Hence, (1-R-T) spectrum may be used as a useful measure of plasmon absorbance peaks for these Mn nano-structures. The spectra obtained by this procedure are given in Figs. 6–8. According to the theoretical results and the model discussed in Section 2, the following may be taken into consideration when analysing these spectra:

- i) The incident angle of the light is an important parameter in exciting plasmon peaks [23,40,41].
- ii) The tilt angle of nano-columns relative to each other should affect the position of the Plasmon peak.
- iii) Plasmonic peaks are related to the interaction of the incident light field with arms of the structure. So if the incident electric field is in the same direction as the arm then the strongest
interaction may occur and if they are normal to each other the weakest interaction will happen\cite{23}.

iv) The position of the plasmonic peak in the extinction spectra depends on the size of the arm: the longer the arm of the structure the longer the wavelength at which the peak occurs and vice versa. For similar lengths, but different radii, the extinction spectrum of the arm with larger radius shows a blue shift. For two similar arms, the more parallel the incident electric field and the arm, then the longer the wavelength of the peak (i.e., by increasing the angle between the electric field and the arm one should observe blue shift)\cite{23}.

v) According to the structure zone model\cite{42–47} grain size (diameter) increases with film thickness. As explained in the experimental section considering the structure with ten arms, the top two arms of this structure are of 36 nm length and the following four arms are of 72 nm length and the last (bottom) four arms are of 110 nm length. Hence, considering the above discussion, the aspect ratio of the top two arms is less than the underlying four arms of the second pitch and the aspect ratio of the latter is again less than the arms of the first pitch. This argument can apply similarly to the structures with 9 and 8 arms.

vi) Siabi and Savaloni\cite{19} by using the homogenization theory reported that the effective metal film thickness interacting with incident light is about 320 nm of the top layers. According to the Tait equation\cite{34} (Eq. (3)) almost eight top arms of the structure with ten arms (with a thickness of 330.5 nm; including two short arms, four medium length arms and two long arms), seven top arms of the structure with nine arms (with a thickness of 310.1 nm; including one short arm, four medium arms and two long arms) and six and a half arms (with a thickness of 321.4 nm; including four medium arms and two and a half long arms) of the structure with eight arms may interact with the incident light electric field.

vii) Siabi and Savaloni\cite{23} observed abnormal behaviour in their results as increased extinction which was found to be due to strong decrease in reflection while the transmission was not affected. They explained this phenomenon in the following way; when the incident light electric field is normal to the first arm (arm on the film surface) it will not interact with this arm

![Image](image.png)

**Fig. 8.** (i) Optical spectra obtained at $\theta=10^\circ$ incident light angle using both $s$- and $p$-polarized lights and different azimuthal angles ($\varphi$) for the graded helical square tower-like (terraced) Mn scupltured thin films produced at different distances (D) from the edge of the shadowing block with 8 arms. (ii) Optical spectra obtained at $\theta=60^\circ$ incident light angle using both $s$- and $p$-polarized lights and different azimuthal angles ($\varphi$) for the graded helical square tower-like (terraced) Mn scupltured thin films produced at different distances (D) from the edge of the shadowing block with 8 arms.
and passes this arm with no disturbance and become trapped in the underlying structure and becomes totally absorbed (i.e., the top layer has an anti-reflection effect).

viii) According to our calculations in Section 2, a blue shift should be considered for two or more parallel nano-rods with different lengths, radii and spacing, while this shift is more pronounced for the case when the number of parallel nano-rods is increased.

Considering the influence of the above mentioned parameters on the extinction spectra and the number of sample sets and number of samples in each set we are forced to investigate the results in different groups/categories so that we can distinguish clearly the influence of each parameter on the plasmonic peaks.

Figs. 6–8 show that in general both s and p spectra obtained for varying distances from the edge of the shadowing block and at varying incidence angles are all different. In general one can mention that the reflectance (absorption) obtained either by using s or p incident polarization increases (decreases) with increasing distance from the edge of the shadowing block. This is due to the reduction of the volume of the voids in the structure of the film with increasing distance from the edge of the shadowing block, as confirmed by both AFM and FESEM results (Table 3).

We discuss the results in order of the incidence angle for both s and p polarizations, for three different distances from the edge of the shadowing block in each set of samples (i.e., samples with 10, 9 and 8 arms) as follows:

a) 10° incidence angle on GHSTS Mn thin film with ten arms:

At 10° angle of incidence, for isotropic films, we would expect very little difference between s and p polarization results since the electric field for both polarisations lies almost in the plane of the film. However, because of the helical square nanostructures in these samples we would expect s-polarization results at azimuthal angle \( \varphi = 0^\circ \) to be identical to p-polarization results at \( \varphi = \pm 90^\circ \). This is clearly seen in Fig. 6(i) for all three samples produced at different distances from the edge of the shadowing block.

As these films are tilted with respect to the substrate normal their arms are also inclined, hence the angle between the electric field (s- or p-polarization) and the arms of these helical square sculptured structures should affect the results. This effect should
be more pronounced for the sample positioned closer to the shadowing block due to its larger tilt angle (Table 3).

Considering the Tait equation (Eq. (3)) for the deposition angle of 80° used in this work the value calculated for \( \beta \) is 55.5°. Hence, the rise angle (\( \chi \)) of each arm relative to the substrate surface is 34.5°. For the A5 sample positioned at 5 cm from the edge of the shadowing block this angle should change due to inclination of the columns of the helical structure. For the structure with ten arms this angle for the four top (first to forth) arms may change as follows: unchanged, increased, unchanged and reduced. This order will repeat for the next four arms and so on. According to the experimental results obtained from the FESEM images the tilt angle for the columns in this sample is 4° (Table 3). Hence, one can obtain the resulting angles for the top eight arms of this structure in which \( \beta \) increases or decreases with the growth angle of helical columns, \( \alpha_r \), as \( \beta + \alpha_r \), or \( \beta - \alpha_r \), as: 55.5° (unchanged), 59.5° (increased) (both are of smaller length (36 nm) and the second one is moved to larger angle: therefore the resulting effect of these two arms is a blue shift). For the next four arms of 72 nm in length the affecting angles are 55.5° (unchanged), 51.5° (decreased), 55.5° (unchanged) and 59.5° (increased) all these arms are of medium length and they may cause a medium shift with respect to the variation of the length. However, with respect to the angle the second arm in this set may cause a red shift while the fourth arm may cause a blue shift and the other two arms have no effect on the shift of the spectrum. The net result is that the influence of the variation of the angle on the extinction spectrum may be cancelled and have no effect. Therefore, we may consider the effect of these four arms as a medium shift in the extinction spectrum. The seventh and eight arms of this structure are of 110 nm length and their angle in the structure is 55.5° (unchanged) and 51.5° (decreased). Their lengths are longer and their radii are smaller than all other arms discussed so far, hence they should cause a red shift, while the angle of the second arm in this set is also decreased which causes a red shift. Hence, on the whole these two arms should red shift the extinction spectrum.

From the above discussion one may deduce that the two short arms produce the shorter wavelength peak (~400 nm) and the four medium (72 nm) length arms may have produced the peak at about 550 nm wavelength and the one or two longer arms (110 nm) should have produced the peak appearing at about 860 nm wavelength for the samples positioned at 5 cm from the edge of the shadowing block. However, considering that the growth angle of
helical columns, $\alpha_t$, increases on decreasing the distance from the edge of the shadowing block (Table 3) the above mentioned effects should become more pronounced on the extinction spectra of A1 and A3 samples which can clearly be seen (Fig. 6(i) b and c).

b) 60° incidence angle on GHSTTS Mn thin film with ten arms

The spectra obtained for s-polarization at 60° incident light angle does not show much change from those obtained at 10° incident light angle other than the intensity of the spectra being reduced. As discussed by Siabi and Savaloni [23] this is due to the closeness of the incident light angle (60°) to the Brewster angle. In the s-polarization spectra of samples positioned at 5 cm and 3 cm from the edge of the shadowing block (Fig. 6(ii) column l(a and b)) a second peak at long wavelengths (around 1000 nm) can be distinguished. This could be the result of interaction of the longer arms with the incident light electric field at this incident light angle [23].

At 60° angle of incidence p-polarization spectra are quite different to those for s-polarization. For samples positioned at 5 cm and 3 cm from the edge of the shadowing block, Fig. 6(ii) (column ll(a and b)) shows that the peak(s) observed before at low wavelengths are shifted to even shorter wavelengths (blue shift) and have almost moved out of the wavelength range of our measurement. The cross over point is also moved in the same direction though is not as distinct as that observed in the spectra of 10° incident light angle. The peak observed at longer wavelengths for 10° incident light angle (~860 nm) is smeared out and one may speculate a blue shift for that peak too observe the hump at about 750 nm wavelength in Fig. 6(ii) column ll(a). All these suggest that the whole spectra have undergone a blue shift. In particular, it can be observed that a new peak is moving inside the wavelength region of our measurement from the right side of the spectrum (long wavelengths). Hence, it can be deduced that at this incident light angle (60°) the p-polarization electric field is either mostly interacting with short arms or its angle of incidence with the effective arms as expected is increased.

c) 10° incidence angle on GHSTTS Mn thin film with nine arms

The similarity in spectra obtained for s- and p-polarized light when the azimuthal angle changes by 90° can again be seen in (Fig. 7(i)). Notably, in some of the spectra of the B1 and B3 samples (i.e., spectra of 45°, 120° and 150° azimuthal angles of the B3 sample and spectrum of 150° azimuthal angle for the B1 sample) the intensity has increased considerably and the spectral shape is also different from the rest of the measurements. The reason for these changes as discussed in the preceding sections was explained by Siabi and Savaloni [23] to be due to a large decrease in reflection in
non-transmitting films, hence extinction is increased. They found that this occurs when the electric field of the incident light is normal (or near normal) to the long axis of the nano-rod. Hence, it may be concluded that for the B3 sample the incident light is near normal to the effective arms of the structure when the azimuthal angle is at 45° (the incident light is normal to the second, forth, sixth and eight arms), 120° and 150° (the incident light is almost normal to the first, third, fifth, seventh and ninth arms). For the B1 sample this situation occurs when the sample is rotated to an azimuthal angle of 150° (the incident light is almost normal to the first, third, fifth, seventh and ninth arms). One should notice the higher intensity of the spectrum obtained for 150° azimuthal angle relative to that of 120° in Fig. 7(i) row I(c) and row II(c) which can be due to the increased angle between the incident light electric field with the long axis of these effective arms. In Fig. 7(i) row I(b) and row II(b) this phenomenon is not observed. This can be due to increased tilt angle of the long axis of the helical square structure (see Table 3) from 8° for the B3 sample to 12° for B1 sample.

Apart from the points mentioned above if one compares the results in Fig. 7(i) (10° incident light angle; 9 arms) with that of Fig. 6(i) (10° incident light angle; 10 arms) an overall red shift in all corresponding spectra can be distinguished.

This red shift in the spectra of B1, B3 and B5 relative to A1, A3 and A5 can be explained in the following way; when the number of arms in the structure is reduced from ten to nine, it means that the last arm of the structure with ten arms (i.e., the top arm with 36 nm length) is missing. Therefore, the top arm of the structure with nine arms is in fact the second arm of the structure with ten arms. If we consider again the effective thickness of the film (~320 nm) which interacts with the incident light electric field then by omission of one short arm at the top (36 nm) the incident light should penetrate further inside this sample and interact with the longer arm at the bottom (arm with 110 nm length and reduced angle between this arm and the electric field of the incident light, both causing a red shift.), hence the interaction with long arms become more dominant and causes a red shift in the spectra.
d) 60° incidence angle on GHSTTS Mn thin film with nine arms

All the spectra in Fig. 7(ii) for both s- and p-polarized incident lights as expected and explained in the preceding sections, show a red shift due to the facts that longer effective arms are involved in interaction with the incident light electric field and there is a decrease of angle between the long axis of arms and the incident electric field. The spectra also show notable increases in intensity at 150° azimuthal angle for the B1 and B3 samples. The consistent occurrence of this intriguing abnormal phenomenon at an azimuthal angle of 150° (which can also be observed for the samples with eight arms see Fig. 8, discussed in the following section) leads us to suggest a structural change in samples which are positioned closer to the shadowing block (and therefore located in the shadow of the block at certain angle of rotation of the sample holder) relative to the one which is positioned out of the shadow of the shadowing block (i.e., at 5 cm distance). The sample positioned at 5 cm from the edge of the shadowing block receives the same amount of adatoms flux at all times during rotation of the substrate holder, therefore the helical square structure is made up of similar arms at each pitch. However, those samples positioned in the shadow of the shadowing block are different since during rotation of the substrate holder when they move into the shadow of the block they do not receive the same amount of adatom flux as when they are out of this shadow. Therefore, the arm formed for the helical square structure in this position is not the same as the other three arms of the structure. This means in the extreme case one can imagine a gap in the structure of the helix in one direction. If this happens then the reflection of the incident light in this direction should decrease as the light should penetrate deeper in the helical structure and because in all cases the films are much thicker than 320 nm (as discussed before [19]) it will get trapped in the film, resulting in higher extinction values as observed in the spectra in Figs. 7 and 8 as abnormal phenomenon.

e) 10° incidence angle on GHSTTS Mn thin film with eight arms

This structure lacks the top two arms of 36 nm length which we have in the structure with ten arms or the top one arm in the structure with nine arms. Hence, the number of parallel effective arms that may interact with the incident light electric field are less, although the arms are now longer and narrower.

According to our model, the DDA calculations in Section 2 and the discussion in the preceding section, longer and narrower arms at the bottom of this structure are involved in the interaction with the incident light, hence a red shift in the obtained spectra is
expected and observed in Fig. 8(i). Overall, the peak that appeared at low wavelengths in the structure with ten arms is shifted to shorter wavelengths in the structure with nine arms and to even longer wavelengths in the structure with eight arms. This is particularly noticeable when the spectra of the samples positioned at 5 cm distance from the edge of the shadowing block are compared in Figs. 6–8. The peak in the spectra of the structure with eight arms is positioned at about 600–700 nm, while that of structure with nine arms occurs at about 500–550 nm and that of the structure with ten arms appears at about 400–450 nm.

In addition, our DDA calculations in Section 2 showed that when the rise angle of the nano-rod (\( \gamma \)) increases, the peak in the spectra moves towards shorter wavelengths (blue shift) and visa versa. This observation can be used in explaining the variation of peak positions in the spectra of the samples located closer to the edge of the shadowing block (i.e., at 1 cm and 3 cm) for samples with ten, nine and eight arms in this work (Figs. 6–8). The structures of the nano-columns formed on these samples are tilted with respect to the substrate surface normal and the tilt angle of these structures for all samples are given in Table 3.

f) 60° incidence angle on GHSTTS Mn thin film with eight arms

Considering the number and the lengths of the arms involved in the interaction of the incident light with this structure mentioned in Section 4.2 (i.e., four of 72 nm length and two and a half of 110 nm length arms (the aspect ratio of half of 110 nm arm is larger than that for a full length of 36 nm arm involved in both structures with nine and ten arms)) one expects that all of these arms cause red shift (or enhance the medium wavelength peak) relative to the structures with nine and ten arms discussed above. The influence of the changing tilt angle (blue shift) produced by decreasing the distance from the edge of the shadowing block can also clearly observed in the spectra of Fig. 8(i) and (ii).
5. Polarization dependent optical spectra

Sculptured thin films produced in this work are of anisotropic nature. Therefore, their anisotropy may be deduced from their extinction spectra as discussed in detail in Section 4. This anisotropy can be seen most clearly in polar plots.

Figs. 9–11 show the angular dependent spectra of the GHSTTS Mn thin films with 10, 9 and 8 arms for both s- and p-polarized incident lights at two incidence angles of 10° and 60° and for different wavelengths, respectively. The intensity at three wavelengths of the extinction spectra were used for plotting these polar figures, namely short, middle and long wavelengths.

The best equation to fit the data is given by:

$$Q_{\text{Ext}} = A + B \sin^2 (\theta + C)$$

(4)
where θ denotes the polarization angle (sample azimuthal angle with respect to the direction of the incident s- or p-polarized light).

The curves in these figures are results of fitting of Eq. (4). The goodness of fit is better than 95%. These polar figures clearly show which of the extinction spectra are anisotropic.

Fig. 9(i) and (ii) show the angular dependent spectra of the structure with 10 arms obtained from 10° and 60° incident light angles (column I: s-polarization and column II: p-polarization), respectively. Almost all of the results shown in these figures including the samples at different distances from the edge of the shadowing block show nearly circular distribution of the polar diagram. This indicates that these structures are almost isotropic and the extinction coefficient does not depend on the azimuthal angle.

In Fig. 10 (i) and (ii) the polar diagrams obtained for the structure with 9 arms are shown. The results obtained for the 10° and 60° incident light angles and for both s- and p-polarizations for the sample at 5 cm from the edge of the shadowing block show circular distributions. This clearly illustrates that at far distance (out of the shadow of the shadowing block), due to arrival of continuous and similar flux of adatoms the structure grows almost isotropically. However, results at 10° incident light angle for the samples positioned at 3 and 1 cm from the edge of the shadowing block for both s- and p-polarizations show oval shaped distributions with an upward lobe at 270° azimuthal angle at short wavelength, which on increasing the wavelength changes to two ovals orthogonal to each other (e.g., a horizontal one and a vertical one). It can also be observed that by increasing the wavelength the horizontal oval decreases in size and the size of the vertical oval increases. These observations indicate that the structure of these samples are anisotropic and strongly depend on the azimuthal angle and at a certain wavelength undergo a phase change. All of these may be due to the abnormal behaviour of these structures which was explained as being a function of the direction of the electric field of the incident light with respect to the axes of the nano-rod(s).

When the incident light angle is increased to 60° (Fig. 10(ii)) the behaviour of the s-polarized light is similar to that obtained at 10° incident light angle (Fig. 10(i) column I) while the behaviour of the p-polarized light for the samples positioned at 1 and 3 cm from the edge of the shadowing block is changed and does not show the strong anisotropy observed at 10° incident light angle. This can be due to the fact that the p-polarized light is normal to the s-polarized light and at 60° incident light angle the s-polarized light remains on the surface of the sample while the p-polarized light penetrates into the structure of the sample almost along the long axis of the helical structure.

Results for the structure with eight arms are fairly similar to those of the structure with nine arms and similar conclusions may be drawn (Fig. 11(i) and (ii)).

The differences observed between the results for the structure with 10 arms (i.e., isotropic at all distances from the shadowing block and at both incident light angles (Fig. 9(i) and (ii))) and those obtained for the structures with 9 and 8 arms (i.e., anisotropic for both s- and p-polarizations at 10° incident light angle and anisotropic for s-polarization at 60° incident light angle for samples positioned at 1 and 3 cm from the edge of the shadowing block) may be explained on the basis of the length of the effective arms on (or near) the surface of these structures. In case of the structure with ten arms one may consider that the two small and fat arms of 36 nm in length (the top two arms) contribute in the structure of the surface of this sample. These arms have angle of 34.5° between them along the z-direction which due to the tilted terraced structure of this helical structure do not create high anisotropy. However, in the case of the structure with 9 arms, the surface includes one small and fat arm of 36 nm and one longer arms of 72 nm which together create the anisotropic structure. If the structure with 8 arms is considered, the surface includes two arms of 72 nm in length at 90° degree angle to each other and 34.5° degree angle in the z-direction to each other. This again causes anisotropy. The results of surface roughness obtained from the AFM analysis of these structures given in Table 2 and discussed in Section 4 also revealed that the surface roughness decreased on increasing the number of arms for the corresponding distances from the edge of the shadowing block. Hence, this should also contribute in the anisotropy and higher anisotropy is expected for the structure with 8 arms relative to the structure with 9 arms and the structure with 9 arms relative to the structure with 10 arms as observed in the polar figures (Figs. 9–11).

6. Conclusions

Graded helical square tower-like (terraced) Mn sculptured thin films with different number of arms were designed and fabricated using oblique angle deposition and a shadowing block at the centre of the substrate holder together with 90° sudden rotations of the substrate holder after completion of each arm. It was observed that the plasmonic peaks obtained from optical measurements at two incident light angles of 10° and 60° and two s- and p-polarizations vary with distance from the shadowing block and depend on structural factors such as lengths and radii of arms (nano-rods), orientation of nano-rods relative to each other and structural defects. Considering that the incident light can only penetrate to a limited thickness (~320 nm) of the structure of the film and the structures fabricated in this work consist of different types of arms (length, radius and orientation) hence different types of shifts in their spectra are observed and identified. Therefore, it is possible to tune the plasmonic peaks by design and engineering of sculptured structures. The proposed model in this work and the use of discrete dipole approximation produced results in good agreement with the experimental observations as well as predicting the structural defects formed in the structure of these sculptured thin films during deposition and growth.

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