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Height and Feature Parameters Study of thermally evaporated ZnS thin films By AFM

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Abstract
AFM has allowed to get microscopic information on the surface structure and to plot topographies representing the surface relief. In this work, this technique was used to visualize the surface relief, specify the growth, and determine Height parameters for form removed thermally evaporated Zinc Sulphide thin films such as root mean square height, Surface Skewness, Surface Kurtosis, Arithmetical mean height , Density of peaks and others. It can be seen on optical micrographs the fine grains with different sizes which are distributed over a smooth homogeneous background that may correspond to the amorphous, or polycrystalline phase of ZnS film. Some of the grains are seen to be united/fused- forming agglomerates. The surface roughness parameters were determined by using the software of ISO 25178 standard provided with the microscope.
Keywords: *Form removed surfaces (FR), roughness (R), waviness (W), root mean square height (RMS), Surface Skewness (Ssk), Surface Kurtosis (Sku), Arithmetical mean height (Sa), Density of peaks and Arithmetic mean peak curvature.*

1. Introduction

Zinc sulphide (ZnS) is an important (II-VI) semiconducting material with a wide direct band gap of 3.65eV in the bulk[1], a high refractive index (2.35) and high dielectric constant[2]. ZnS thin films with a wide direct band gap and n- type conductivity are promising candidates for optoelectronic device applications. These properties of zinc sulphide thin films are promising for short wavelength optoelectronic device applications. It has potential applications in optoelectronic devices such as blue light emitting diodes[3] ZnS can also be used for light emitting diodes in the blue ultraviolet region because of its wide band gap. ZnS is also widely used as the base materials for cathode-ray tube luminescent materials, catalysts, electroluminescent devices, and UV semiconductor lasers for optical lithography [4,5]. ZnS crystals are material having high photoluminescence and thermo –luminescence properties above room temperature. The nanostructures made up of ZnS materials find attractive applications in electronic and optoelectronic nano-devices [5],electroluminescent devices and photovoltaic cells[6]. In thin film solar cells based on CuGaIn (S.Se)2 absorbers, a CdS buffer layer is generally required in order to obtain high conversion efficiency. However, there are toxic hazards with respect to the production and use of the CdS layer. Therefore research in developing Cd-free buffer layers has been encouraged. This has lead to the investigation of ZnS as a buffer layer in ZnO/ZnS/CuInS2 devices[7]. ZnS has a wider energy band gap than CdS, which results in the transmission of more high energy photons to the junction and to the enhancement of the blue response to the photovoltaic cells.

Several techniques such as thermal evaporation [8], molecular beam epitaxy [9], metal-organic vapor phase epitaxy [10], chemical vapor deposition[11], spray pyrolysis[12], and chemical bath deposition CBD[13] have been used to produce ZnS thin films for photovoltaic applications because of its efficient, cost effective and large scale capability[15].

2. Theoretical study of roughness and surface parameters [16, 17, 18]

The Roughness of a surface is defined as the main components of the relief containing the smallest wavelengths measured on the sample. The roughness gives indication on the nature of the material and the machining type used. The wavelengths under the cut-off are kept in the roughness, the others, higher, are kept in the waviness.

The height properties, according to surf are described by six parameters, which give information about the statistical average properties, the shape of the height distribution histogram and about extreme properties. All the parameters are based on two-dimensional standards that are extended to three dimensions.

A. Height Parameters (ISO 25178) (Surface)

Parameters related to roughness are general and valid for any M N rectangular image.

Some of the parameters depend on the definition of a local minimum and a local maximum. Here, a local minimum is defined as a pixel where all eight neighboring pixels are higher and a local maximum as a pixel where all eight neighboring pixels are lower.

As there are no pixels outside the borders of the image there are no local minimums or local maximums on the borders. Note that parameters based on local minimums and/or local maximums may be more sensitive to noise than other parameters.

Height parameters are a class of surface finish parameters that quantify the Z-axis perpendicular to the surface. They are included in the ISO 25178 standard.

The reference plane for the calculation of these parameters is the mean plane of the measured surface.

1. Root mean square height, S_q

Standard deviation of the height distribution, or RMS surface roughness. It is defined as:

$$S_q = \sqrt{\frac{1}{MN}\sum_{k=0}^{M-1}\sum_{l=0}^{N-1}[z(x_k, y_l)]^2}$$

Computes the standard deviation for the amplitudes of the surface (RMS).

2. The Surface Skewness or skewness of the height distribution, S_{sk}, is defined as:

$$S_{sk} = \frac{1}{MN\ S_q^3}\sum_{k=0}^{M-1}\sum_{l=0}^{N-1}[z(x_k, y_l)]^3$$

which is the third statistical moment, qualifying the asymmetry of the height distribution histogram. A negative S_{sk} (Fig. 1a) indicates that the surface is composed with principally one plateau and deep and fine valleys. In this case, the distribution is sloping to the top. A positive S_{sk} (Fig. 1b) indicates a surface with lots of peaks on a plane. The distribution is sloping to the bottom. Due to the big exponent used, this parameter is very sensitive to the sampling and to the noise of the measurement.

If S_{sk} = 0, a symmetric height distribution is indicated, for example, a Gaussian like. If $S_{sk} < 0$, it can be a bearing surface with holes and if $S_{sk} > 0$ it can be a flat surface with peaks. Values numerically greater than 1.0 may indicate extreme holes or peaks on the surface.

3. The Surface Kurtosis or Kurtosis of the height distribution, S_{ku}, is defined as:

$$S_{ku} = \frac{1}{MN\ S_q^4}\sum_{k=0}^{M-1}\sum_{l=0}^{N-1}[z(x_k, y_l)]^4$$

Which is the fourth statistical moment, qualifying the flatness of the height distribution. Due to the big exponent used, this parameter is very sensitive to the sampling and to the noise of the measurement.

For Gaussian height distributions S_{ku} approaches 3.0 when increasing the number of pixels. Smaller values indicate broader height distributions and *visa versa* for values greater than 3.0.

A high kurtosis distribution has a sharper *peak* and longer, fatter *tails*, while a low kurtosis distribution has a more rounded peak and shorter thinner tails.

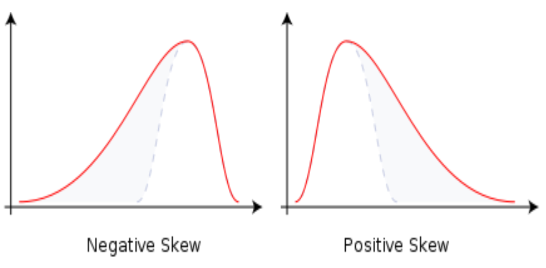

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Figure 1.

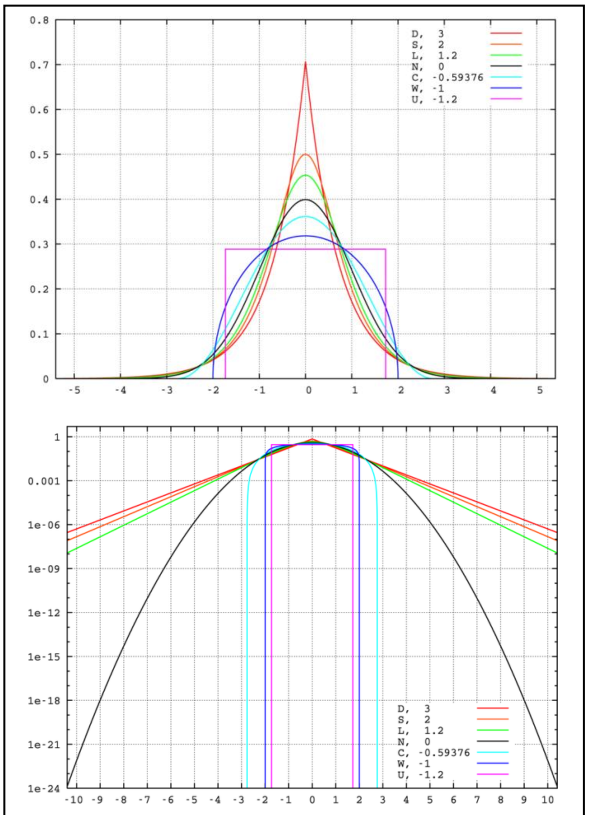


Figure 2. In this example we compare several well-known distributions from different parametric families.

4. Maximum peak height, S_p, Height between the highest peak and the mean plane.

5. Maximum pit height, S_v, Depth between the mean plane and the deepest valley.

6. Maximum height, S_z

Height between the highest peak and the deepest valley.

The definition of the (ISO 25178) Sz parameter is different from the definition of the (EUR 15178N) Sz parameter. The value of the (EUR 15178N) Sz parameter is always smaller than the value of the (ISO 25178) Sz parameter.

The (ISO 25178) Sz parameter replaces the (EUR 15178N) St parameter.

The **Peak-Peak Height,** are denoted by three parameter names, ***S_z, S_t, S_y,*** according to ISO, ASME and reference [6]. They are defined as the height difference between the highest and lowest pixel in the image.

$$S_z = S_t = S_y = z_{\max} - z_{\min}$$

7. Arithmetical mean height, S_a or Mean surface roughness is defined as

$$S_q = \frac{1}{MN}\sum_{k=0}^{M-1}\sum_{l=0}^{N-1}|\,z(x_k, y_l)\,|$$

B. Theoretical study of Feature parameters [16, 17, 18]

The feature parameters are a new family of parameters that is integrated in the ISO 25178 standard. Feature parameters are derived from the segmentation of a surface into motifs (hills and dales). Segmentation is carried out in accordance with the watersheds algorithm

For the moment, all feature parameters are calculated after a discrimination by segmentation using a Wolf pruning of 5% of the value of the Sz parameter (Maximum height).

1. Density of peaks, S_{pd}, Number of peaks per unit area.

$$S_{pd} = \frac{Number\ of\ local\ Maximums}{(M-1)(N-1)\delta x \delta y}$$

Because, the parameter is sensitive to noisy peaks it should be interpreted carefully.

The peaks taken into account for the (EUR 15178N) S_{ds} parameter are detected by local neighbourhood (with respect to 8 neighbouring points) without discrimination between local and significant peaks. The (ISO 25178) S_{pd} parameter is calculated the same way, but takes into account only those significant peaks that remain after a discrimination by segmentation (Wolfpruning of 5% of Sz). Therefore the value of the (ISO 25178) S_{pd} parameter is smaller than the value of the (EUR 15178N) S_{ds} parameter.

2. Arithmetic mean peak curvature, S_{pc} (Mean Summit Curvature, S_{sc}, according to SPIP) Arithmetic mean of the principle curvatures of peaks within a definition area. It is defined as:

$$S_{pc} = \frac{-1}{2n}\sum_{i=1}^{n}\left(\frac{\delta^2 z(x,y)}{\delta x^2}\right)+\left(\frac{\delta^2 z(x,y)}{\delta y^2}\right)$$

for all local maximums , where δx and δy are the pixel separation distances. This parameter enables to know the mean form of the peaks: either pointed, either rounded, according to the mean value of the curvature of the surface at these points.

The peaks taken into account for the (EUR 15178N) S_{sc} parameter are detected by local neighbourhood (with respect to 8 neighbouring points) without discrimination between local and significant peaks. The (ISO 25178) S_{pc} parameter is calculated the same way, but takes into account only those significant peaks that remain after a discrimination by segmentation (Wolf pruning of 5% of S_z). Therefore the value of the (ISO 25178) S_{pc} parameter is more accurate and significant than the value of the (EUR 15178N) S_{sc} parameter.

There are three hybrid parameters. These parameters reflect slope gradients and their calculations are based on local z-slopes.

3. Ten point height, S_{10z}. Average value of the heights of the five peaks with the largest global peak height added to the average value of the heights of the five pits with the largest global pit height, within the definition area.

$$S_{10z} = S_{5p} + S_{5v}$$

or $$S_{10z} = \frac{\sum_{i=1}^{5}|z_{pi}| + \sum_{i=1}^{5}|z_{vi}|}{5}$$

where z_{pi} and z_{vi} are the height of the i^{th} highest local maximums and the i^{th} lowest local minimums respectively. When there are less than five valid maximums or five valid minimums, the parameter is not defined.

4. Five point peak height, S_{5p}. Average value of the heights of the five peaks with the largest global peak height, within the definition area.

5. Five point pit height, S_{5v}. Average value of the heights of the five pits with the largest global pit height, within the definition area.

6. Closed dale area, S_{da}. Mean dale area Average area of dales connected to the edge at height c.

7. Closed hill area, S_{ha}. Mean hill area Average area of hills connected to the edge at height c.

8. Closed dale volume, S_{dv}. (Mean dale volume) Average volume of dales connected to the edge at height c.

9. Closed hill volume, S_{hv}.(mean hill volume) Average volume of hills connected to the edge at height c.

3. Experiments and AFM Characterization

Thin films of ZnS films were deposited on glass substrates by thermal evaporation. Experimental evidence and analytical studies of the XRD patterns and optical characterizations of ZnS films with different optical thicknesses had been reported previously [19]. In this work, I used AFM technique to get microscopic information on the surface structure, to plot topographies representing the surface relief , specify the growth, and determine the contribution of optical thickness to the quality of the film. The study of height parameters was carried out by contact and dynamic modes of AFM. The optical photos of 3different thicknesses, whose sizes are of (500µm*370µm), show, high in-homogeneity of crystallite sizes. The thicker the sample, the larger the crystallite size is. Fig.3 shows an optical micrograph of zinc sulphide thin films of 300nm thickness, grown on glass substrate.

Before the calculation of the roughness parameters, I carried out a slope correction by a 2^{nd} order polynomial plane fit. Components of the surface relief of long horizontal wavelengths have been removed in order to analyze surface finish (i.e. waviness and roughness). As the Form in our samples is coming from a smooth material (thin films), 2^{nd} order polynomial plane fit Form Removal operator has been used for slope correction. **Gaussian Filtering (**with 0.25 µm cut-off wavelength**) has been used to** separate data frequencies (or wavelengths) into two parts, the first one having the long wavelengths (waviness), the other one having the short wavelengths (roughness).

The wavelengths above the cut-off are kept in the waviness whereas the others, smaller, are kept in the roughness.

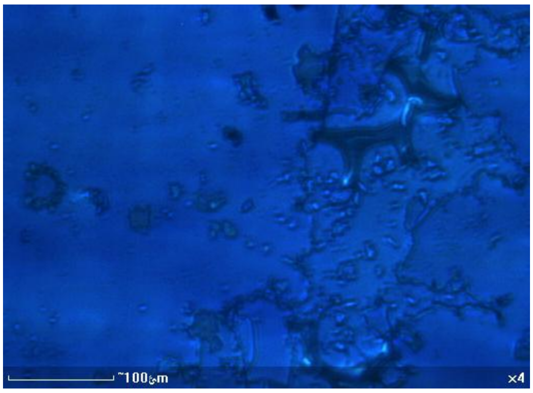
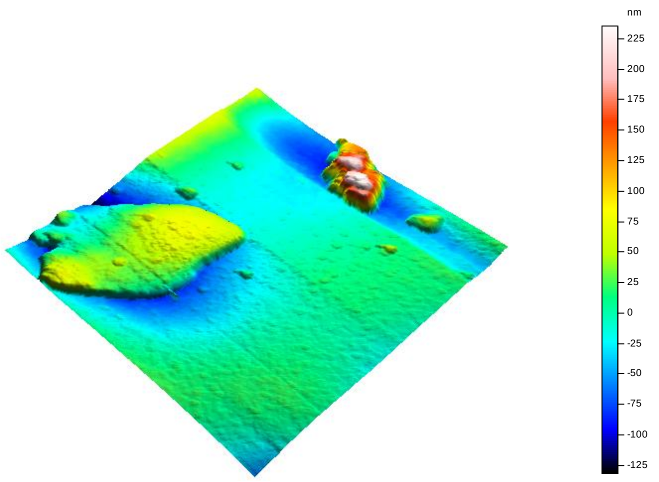


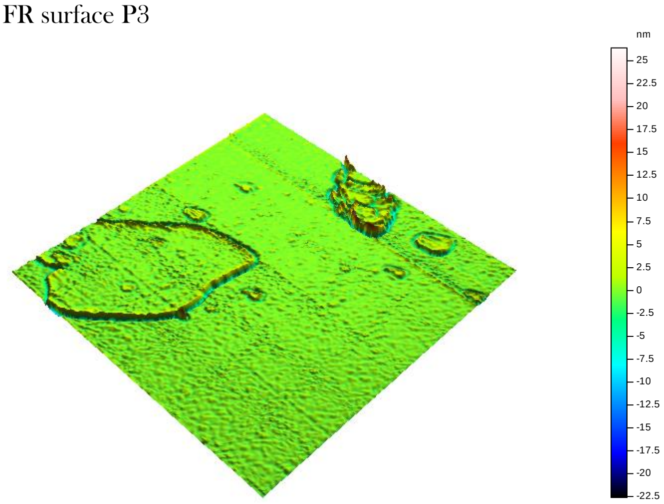
Figure 3. An optical micrograph for (500µm*370µm) area of thermally evaporated ZnS film, of 300nm thickness, grown on glass substrate.

Figs.4,5,6 illustrate the topographies of zinc sulphide thin films of 300, 200, 50nm thickness respectively. Each figure contains 3D AFM view of denoised, form removed surface, and Gaussian filtered roughness for one point of each film.

The table in each figure contains the Height Parameters of form removed (FR) surface, it's roughness and waviness in several points of the concerned film.



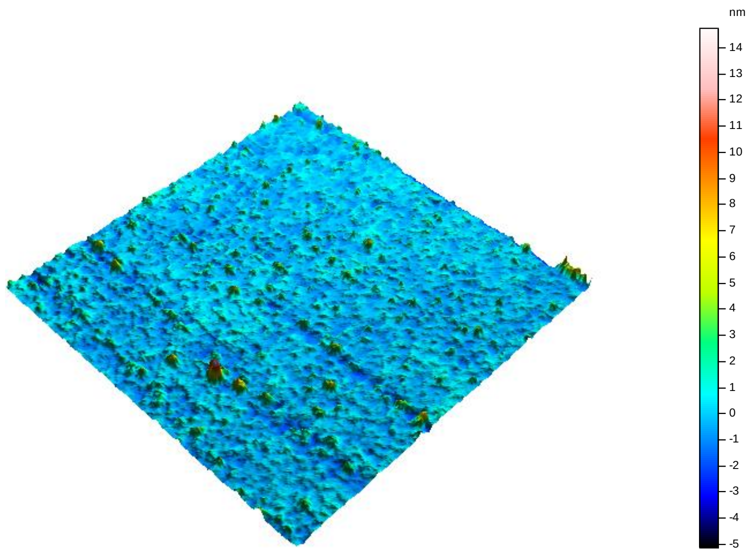
FR surface P3



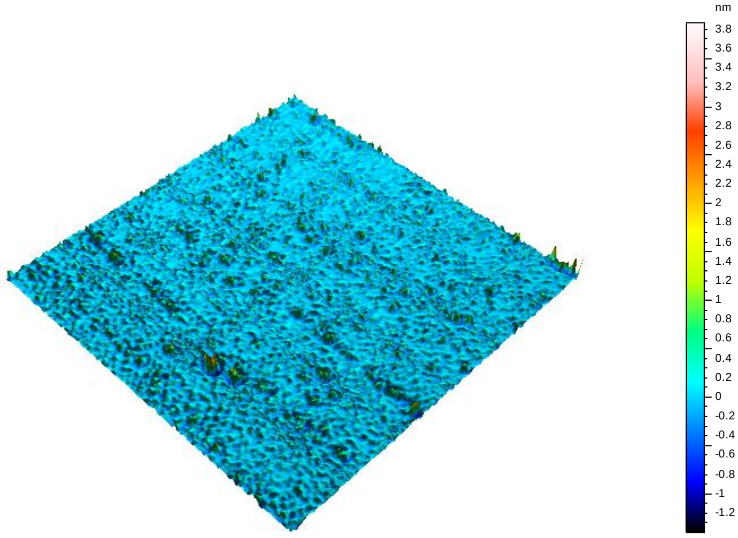
Gaussian filtered roughness P3

ISO 25178				
Height Parameters				
300nm thickness				
		FR	R	W
S_q	P1	113 nm	3.95 nm	0.111 µm
	P2	2.56nm	0.385nm	2.24 nm
	P3	41.4 nm	1.51 nm	40.8 nm
	P4	2.86 nm	0.433nm	0.433 nm
S_{sk}	P1	-0.122	-1.32	-0.123
	P2	6.15	0.466	7.12
	P3	1.47	-1.28	1.49
	P4	5.9	0.57	0.57
S_{ku}	P1	4.9	45.3	4.01
	P2	75.7	13.9	95.2
	P3	8.66	40.1	8.71
	P4	75	8.73	8.73
S_p	P1	443 nm	68.8 nm	0.361µm
	P2	39.9 nm	4.77 nm	39.1 nm
	P3	235 nm	26.4 µm	0.233 µm
	P4	45.5 nm	4.85 nm	4.85 nm
S_v	P1	518 nm	66.7 nm	0.499µm
	P2	8.99 nm	5.54 nm	8.47nm
	P3	132 nm	22.6 nm	0.128 µm
	P4	9.79 nm	4.17 nm	4.17 nm
S_z	P1	961 nm	135 nm	0.86µm
	P2	48.9 nm	10.3 nm	47.6 nm
	P3	367 nm	49 nm	0.362 µm
	P4	55.3 nm	9.02 nm	9.02 nm
S_a	P1	81 nm	1.63 nm	0.0802µm
	P2	1.42 nm	0.275 nm	1.15 nm
	P3	28.4 nm	0.000705	0.028 µm
	P4	1.63 nm	0.321 nm	0.321 nm

Figure 4. 3D view of denoised, form removed surface and Gaussian filtered of AFM photo for point P3 of 300nm thickness thin film of ZnS. The Table indicates the Height Parameters of form removed (FR) surface, roughness (R) and waviness (W) in four points (P1, P2, P3, P4) of the 300nm thickness film.



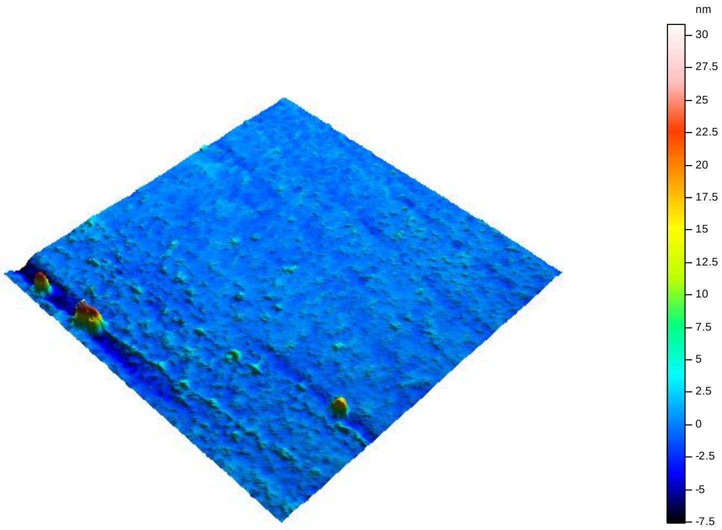
FR surface P4



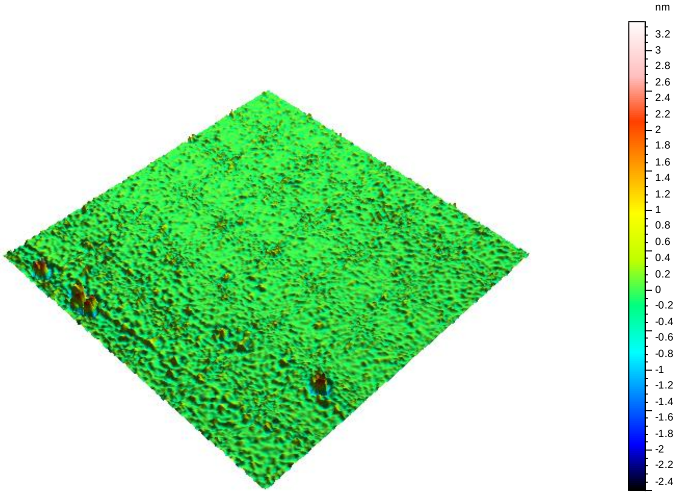
Gaussian filtered roughness P4

ISO 25178				
Height Parameters				
200nm				
		FR	R	W
S_q	P1	3.1 nm	0.369 nm	2.87 nm
	P2	1.56 nm	0.362 nm	1.19 nm
	P3	10.9 nm	0.402 nm	10.6 nm
	P4	0.845 nm	0.193 nm	0.65 nm
S_{sk}	P1	8.41	0.267	9.51
	P2	1.06	0.584	1.04
	P3	12.7	1.6	12.7
	P4	3.02	1.35	3.3
S_{ku}	P1	119	12.6	143
	P2	5	4.03	5.69
	P3	253	184	254
	P4	30.1	15.2	37.3
S_p	P1	52.9 nm	3.63 nm	52.3 nm
	P2	9.79 nm	2.43 nm	8.49 nm
	P3	238 nm	13.3 nm	236 nm
	P4	14.7 nm	3.87 nm	12.5 nm
S_v	P1	8.24 nm	5.04 nm	7.63 nm
	P2	3.86 nm	1.48 nm	3.26 nm
	P3	53.9 nm	11.4 nm	52.3 nm
	P4	5.13 nm	1.4 nm	3.09 nm
S_z	P1	61.1nm	8.67 nm	60 nm
	P2	13.7 nm	3.91 nm	11.7 nm
	P3	292 nm	24.7 nm	288 nm
	P4	19.9 nm	5.27 nm	15.6 nm
S_a	P1	1.51 nm	0.253 nm	1.28 nm
	P2	1.19 nm	0.276 nm	0.903 nm
	P3	3.61 nm	0.162 nm	3.55 nm
	P4	0.551 nm	0.135 nm	0.425 nm

Figure 5. 3D view of denoised, form removed surface and Gaussian filtered of AFM photo for point P4 of 200nm thickness thin film of ZnS. The Table indicates the Height Parameters of form removed (FR) surface, roughness (R) and waviness (W) in four points (P1, P2, P3, P4) of the 200nm thickness film.



FR surface P2



Gaussian filtered roughness P2

ISO 25178				
Height Parameters				
50nm				
		FR	R	W
Sq	P1	4.42 nm	0.488 nm	4 nm
	P2	1.55 nm	0.201 nm	1.39 nm
	P3	2.36 nm	0.33 nm	2.16 nm
Ssk	P1	5.94	-0.918	6.53
	P2	7.79	1.36	8.18
	P3	13.7	0.278	14.1
Sku	P1	83.4	46	94.7
	P2	115	29	124
	P3	243	222	256
Sp	P1	77.3 nm	8.76 nm	73 nm
	P2	30.8 nm	3.36 nm	28.5 nm
	P3	51.8 nm	10.9 nm	50.4 nm
Sv	P1	23.5 nm	8.33 nm	20.9 nm
	P2	7.58 nm	2.51 nm	7.21 nm
	P3	5.79 nm	10.7 nm	5.17 nm
Sz	P1	101 nm	17.1 nm	93.9 nm
	P2	38.4 nm	5.87 nm	35.7 nm
	P3	57.6 nm	21.5 nm	55.5 nm
Sa	P1	1.78 nm	0.241 nm	1.59 nm
	P2	0.699 nm	0.129 nm	0.587 nm
	P3	0.746 nm	0.144 nm	0.632 nm

Figure 6. 3D view of denoised, form removed surface and Gaussian filtered of AFM photo for point P2 of 50nm thickness thin film of ZnS. The Table indicates the Height Parameters of form removed (FR) surface, roughness (R) and waviness (W) in three points (P1, P2, P3) of the 50nm thickness film.

4. Discussion & Conclusion

Investigation of the Gaussian filtered roughness Height Parameters of form removed surfaces for 3 different thicknesses of ZnS film shows that they depend highly the film thickness, and even they vary from point to the other on the same film. The root mean square (RMS) S_q of the roughness on the 300nm thickness film is positioned between 0.00395 µm and 0.385nm, (table of Fig4), between 0.402 nm and 0.193nm (table of Fig5) and between 0.488 nm and 0.201nm (table of Fig6) in the scanned range areas.
Investigating skewness **height distribution,** S_{sk} values in tables of Figs 4, 5, 4 shows that each of the films may include areas with positive surface skewness and others with negative surface skewness, indicating that surface is composed of peaks and holes on a plane. Though the parameter is very sensitive to sampling I didn't notice a big difference between the examined areas.
Examining Kurtosis of the height distributions, S_{ku} in the tables mentioned above, indicates that they are in most cases much greater than 3, indicating sharp peaks and long tails especially in the 50 nm thick film, where surface Kurtosis values are positioned between 29 and 222.
Examining the Density of peaks, S_{pd} of the roughness surface on the 300nm thickness film in the table below, we notice that it is positioned between 518387 1/µm2 and 8620587 1/µm2, for the 200nm thickness between 124561 1/µm2 and 11948300 1/µm2 , and for 50nm between 182051 1/µm2 and 3966797 1/µm2 in the scanned range areas.
Regarding the **Arithmetic mean peak curvature,** S_{pc} values, the table shows that they vary from 0.496 1/µm to 9.01 1/µm(for300nm), from 0.298 1/µm to 1.71 1/µm(for200nm) and from 0.314 1/µm to 1.65 1/µm (for50), which indicates

the sensitivity of this parameter to film thickness, at least in the examined areas.

Values of Height and Feature parameters of ZnS thin films should be controlled according to the application sought after, transmission or reflection or fluid retention.

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