ATOMIC FORCE MICROSCOPY CHARACTERIZATION
OF ZnTe EPITAXIAL FILMS

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In this paper results of a characterization of the surfaces of ZnTe epitaxial thin films exhibiting the different thicknesses are presented. The results mentioned are obtained using the procedures enabling us to determine the values of the following quantities: mean grain size, grain size distribution, root–mean square values of the heights of the irregularities and the diagram describing the distribution of the directions of the normals. For the analysis of the grain structure a watershed algorithm is used. It is shown that the values of these quantities can describe the morphology of the ZnTe film surfaces in a sufficient way. Further, it is shown that the structure of the surfaces of the ZnTe films exhibit facets forming a grain structure. Moreover, it is presented that the ZnTe film surfaces exhibit a strong slope anisotropy and that the linear dimensions of the grains increase with increasing values of the thicknesses of the ZnTe films.

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1 Introduction

Atomic force microscopy (AFM) is a very useful tool for investigating the surfaces (the upper boundaries) of thin solid films. This technique has been used to study a structure of surfaces of films prepared by different technologies. For example, AFM has been employed for analyzing the roughness [1–3], grain structure [4] and fractal properties [5,6] of the thin film surfaces. Further, it has been employed for the thin films formed by different materials, e.g. the films formed by single crystal of semiconductors, columnar films formed by dielectrics, polycrystalline films of metals etc. In this paper a grain structure of the ZnTe epitaxial film surfaces will be studied. In our earlier investigation, we have namely found that the surfaces of these films exhibited a relatively complicated structure that can be understood as a certain kind of the grain structure [7]. The grain structure of the ZnTe films exhibits a pronounced anisotropy, i.e. in different directions within the mean plane of the surfaces the one–dimensional distribution of the slopes is different.

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However, so far no detailed attention has been devoted to this effect and our results are first ones. Therefore, the procedures of analyzing the forms and dimensions of the grains together with results obtained using them will be presented. Moreover, the mentioned anisotropy of the samples will be studied in detail.

2 Experimental arrangement

The epitaxial films of ZnTe were prepared by molecular beam epitaxy (MBE) on (100) GaAs single crystal substrates. The temperature of the substrates was of 300°C. The flux of Zn and Te atoms was monitored with a reference gauge, placed close to the sample holder. The beam equivalent pressure for Zn and Te was \(1 \text{–} 2 \times 10^{-7} \text{ Torr}\) and \(4 \text{–} 16 \times 10^{-7} \text{ Torr}\), respectively. The growth rate was between 4–8 nm/min. We investigated five samples of the ZnTe epitaxial thin
films characterized with the different values of the thicknesses (samples Nos. 1–5). The values of these thicknesses were as follows: $d_1 = 10.8$ nm (No. 1), $d_2 = 96.9$ nm (No. 2), $d_3 = 311$ nm (No. 3), $d_4 = 388$ nm (No. 4), $d_5 = 798$ nm (No. 5). The values of the thicknesses were determined using optical measurements.

For the analysis of the ZnTe films the commercial apparatus ThermoMicroscopes Explorer was used. Both contact and non-contact modes were used for the measurements. Both the standard contact (apex ratio 1:2, apex curvature $< 20$ nm) and non-contact (apex ratio 1:10, apex curvature $< 10$ nm) tips were utilized.

### 3 Data processing

In Fig. 1a the AFM image of the part of the surface of the chosen ZnTe film (sample No. 5) is introduced. From this figure one can see that the structure of this sample corresponds to a system of facets forming the objects resembling the grain structure. Therefore the following analysis of the structure of the ZnTe film surfaces will be performed under assumption that the objects mentioned above are identical with certain grains. This opinion is supported by the fact that the ZnTe epitaxial films exhibit in their volumes a mosaic structure formed by a network of low angle dislocations [8]. Thus, as a first approximation, it is possible to believe that in the ZnTe film surfaces some grains corresponding to the mosaic blocks take place. It should be noted that the aim of this work has not consisted in the study of a relationship between the mosaic structure on the one hand and the grains observed on the other hand. Our main aim consists in finding efficient procedures enabling us to carry out a complete analysis of this kind of the surface structure of ZnTe films using AFM.

For the analysis mentioned we examined the following procedures:

1. grain analysis by both the height and second derivative thresholding,
2. grain tops identification by modified watershed algorithm,
3. determination of the grain geometry using watershed algorithm,
4. mean grain size and grain size distribution evaluation,
5. procedure for creating a diagram describing the distribution of the directions of the normals to the boundaries,
6. procedure for evaluating the root mean square value of the heights.

The first four items are connected with the grain properties of the surface, the last one is connected with the slopes of the facets forming the grains. In the following the algorithms used will be discussed briefly.

The grain analysis by the height thresholding is the simplest and mostly used method for determination of grain positions and volumes. The algorithm is based on selecting a height threshold value (usually at the value of the mean plane of the surface). The parts of the surface that are above this value are denoted as grains, the parts that are below the threshold are denoted as pores. Then the grain statistical properties (grain mean size, grain size distribution etc.) can be evaluated. This algorithm is sufficient one to describe the grain properties of isolated particles.
Fig. 2. Grain identification by the watershed algorithm (first step of the algorithm): A, water drop stops in the local minimum and fills it by its volume, B, when the small local minimum is filled, other drops can pass through it to a larger local minimum.

Fig. 3. Watershed algorithm image segmentation (second step of the algorithm): the numbers denote particles in four possible model positions (see text).

deposited on the planar surface [4]. However, for more complicated grain–like structures (e.g. the columnar structures of metals formed by vacuum evaporation) it fails. The results of the grain analysis of the ZnTe epitaxial films by means of this algorithm are plotted in the Fig. 1b. We can see that the grains are not identified properly: on one side many of the larger grains are merged together on the other side the small grains are not found at all.

The grain analysis by the second derivative thresholding is a modification of the height thresholding method. Here the Laplacian of Gaussian filter is applied on the data. At each point in the array resulting from this filtering the value is thresholded again. In the ideal case this method should clearly identify the tops of the grains as their curvature is different from the curvature of the pores. However, it is clear that this method is very sensitive to noise. The Gaussian filtering embedded into the Laplacian of Gaussian filter is not sufficient to eliminate all the noise in the AFM data. Therefore one must apply some strong filtering to denoise the AFM data which can lead to loss of information about grains. The grains identified by this method are plotted in the Fig. 1c. We can see that the results are better than the results obtained using the height thresholding algorithm but still insufficient for our purposes.

The watershed algorithm is usually employed for local minima determination and image segmentation in image processing [9, 10]. As the problem of determining the grain positions can be understood as the problem of finding local extremes on the surface we used this algorithm for our purposes. For convenience in the following we will treat the data inverted in the $z$ direction while describing the algorithm (i.e. the grain tops are forming local minima in the following text). We applied two stages of the grain analysis:

1. At each point of the inverted surface the virtual water drop was placed. In the case that the drop was not already in a local minimum it followed the steepest descent path to minimize its potential energy. As soon as the drop reached any local minimum it stopped here and rested on the surface. In this way it filled the local minimum partially by its volume (see Fig. 2 and the figure caption). This process was repeated several times. As the result a system of lakes of different sizes filling the inverted surface depressions was obtained. Then the area of each of the lakes was evaluated and the smallest lakes were removed under assumption that they were formed in the local minima originated by noise. The larger lakes were used to identify the positions of the grains. In this way the noise in the AFM data was eliminated.
2. The grains found in the step 1 were marked (each one by a different number). The water drops continued in falling to the surface and filling the local minima. As the grains were already identified and marked after the first step, the next five situations could happen as soon as the drop reached a local minimum (see Fig. 3):

- the drop reached the place previously marked as a concrete grain. In this case the drop was merged with the grain (i.e., it was marked as a part of the same grain, see Fig. 3 — particle No. 1),
- the drop reached the place where no grain was found but a concrete grain was found in the closest neighborhood of the drop. In this case the drop was merged with the grain again (see Fig. 3 — particle No. 2),
Fig. 5. Distribution functions of the grain radii for the ZnTe samples Nos. 2–5.

Fig. 6. Diagram of the distribution of the normal components of the surface corresponding to sample No. 5 (the scale of the figure corresponds to the interval from $-18$ to $18$ degrees).

- the drop reached the place where no grain was found and no grain was found even in the closest neighborhood of the drop. In that case the drop was not marked at all (see Fig. 3 — particle No. 3),
- the drop reached the place where no grain was found but more than one concrete grain was found in the closest neighborhood (e.g. two different grains were found in the neighborhood). In this case the drop was marked as the grain boundary (see Fig. 3 — particle No. 4),
- the drop reached the place marked as grain boundary. In this case the drop was marked as the grain boundary too.

After filling all the depressions we obtained a mask of the grain boundaries that determined the grain positions and sizes.

In this way we can identify the grain positions and then determine the volume occupied by each grain separately. In Fig. 1d the grains identified for the ZnTe sample No. 5 (after first few iterations of the second step of the watershed algorithm) are plotted. As a result of the finished watershed algorithm we obtain the image segmented into the grains (see Fig. 4). It can be clearly seen that the results are much better than the results of the other two methods. Therefore, for the evaluation of the statistical properties of the grain structure we used the watershed algorithm in this work.

The mean grain size was determined as follows: a family of the grains was selected. Then the area occupied by every grain was evaluated. A nominal circle having the same area as the grain was assigned to this grain. The radius of this nominal circle was considered to be the size
Tab. 1. The values of the film thicknesses, RMS values of the heights $\sigma$ and grain radii for the samples studied.

<table>
<thead>
<tr>
<th>Sample No.</th>
<th>$d$ [nm]</th>
<th>$\sigma$ [nm]</th>
<th>grain radius [nm]</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>10.758±0.021</td>
<td>0.79±0.05</td>
<td>—</td>
</tr>
<tr>
<td>2</td>
<td>96.875±0.026</td>
<td>1.51±0.05</td>
<td>95±5</td>
</tr>
<tr>
<td>3</td>
<td>311.058±0.037</td>
<td>4.23±0.07</td>
<td>119±4</td>
</tr>
<tr>
<td>4</td>
<td>387.728±0.033</td>
<td>5.95±0.10</td>
<td>209±3</td>
</tr>
<tr>
<td>5</td>
<td>797.596±0.040</td>
<td>13.22±0.15</td>
<td>297±3</td>
</tr>
</tbody>
</table>

of the grain selected. The mean value calculated on the basis of all the grain sizes represented this mean grain size. The family of the values of the size of all the grains selected represented the grain size distribution.

The diagram describing the distribution of the directions of the normals was obtained in the following way: At each point of the AFM scan the normal components to the surface were calculated. This components determined one point in the plane of the diagram. In this point the value was incremented by number one. Then the total number of the increments in the chosen point in the plane of the diagram represented the number of the data points exhibiting the chosen values of the normal components. A normalization of the numbers in the diagram was finally performed.

The root-mean square value of the heights (RMS roughness) was evaluated by means of the formula presented in Ref. 2.

4 Results and discussion

In Fig. 4 the AFM images of the parts of the five ZnTe films studied are plotted together with the masks of the grain boundaries obtained by means of the watershed algorithm. From this figure we can see that both the grain lateral and height dimensions increase with increasing the thickness of the ZnTe film. Note that the grain–like faceted structure was not observed for the thinnest ZnTe film studied ($d_1 = 10.7$ nm). From Figs. 1 and 4 we can see that the watershed algorithm is able to determine correctly the grain positions and boundaries without the need of strong data filtering. Thus, it can be seen that the watershed algorithm can be used very effectively for the AFM data segmentation into separate grains.

In Fig. 5 the distributions of the grain sizes are plotted for all the samples studied. We can see that if the value of the ZnTe film thickness increases, the distribution of the grain sizes gets wider (i.e. more grains with mutually different sizes appear to be on the surface). In Table 1 the values of the thicknesses, mean grain sizes and RMS values of heights are introduced. We can see that both the mean grain size and RMS value of heights increase with the increasing thickness of the ZnTe film. The similar increase of the mosaic block size with the increasing thickness of the ZnTe film is described in the literature concerning X-ray studies of ZnTe samples prepared by molecular beam epitaxy [11]. Unfortunately, for our samples it was very problematic to perform the similar X-ray measurements as described in Ref. 11 due to the small thickness of the samples. The comparison of our results with the X-ray ones will be discussed in our forthcoming paper.
In Fig. 6 the diagram describing the distribution of the directions of the normals is plotted. The preferred directions on the surface (i.e. the anisotropy) can be clearly seen. These directions correspond to the facets forming the grain structure. The facet angles can be evaluated from the distribution too: their values were found to be between 4 and 11°. The facets therefore probably correspond to some planes with relatively high values of the Miller indices.

5 Conclusions

In this paper the procedures enabling us to characterize the surface morphology of the ZnTe films were described in detail. The procedures for the determination of the following quantities were namely presented: mean grain size, grain size distribution, RMS values of the heights and the diagram describing the distribution of the directions of the normals. These quantities can be used to characterize the ZnTe surfaces in an efficient way (mainly the grain and facet structure are important for characterizing these surfaces). Using the interpretation of the quantities mentioned above it was found that the ZnTe surfaces exhibited the strong anisotropy in the slopes of the facets. Further, it was found that within the surfaces the facets formed the grain-like structures. The linear dimensions of these structures increased with the increasing values of the thicknesses of the ZnTe films under study. Thus, it was shown that AFM is the suitable technique for characterizing the ZnTe film surfaces structure. Of course, it is evident that the same analysis can be appropriate for characterizing the other thin films exhibiting a similar surface structure.

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References