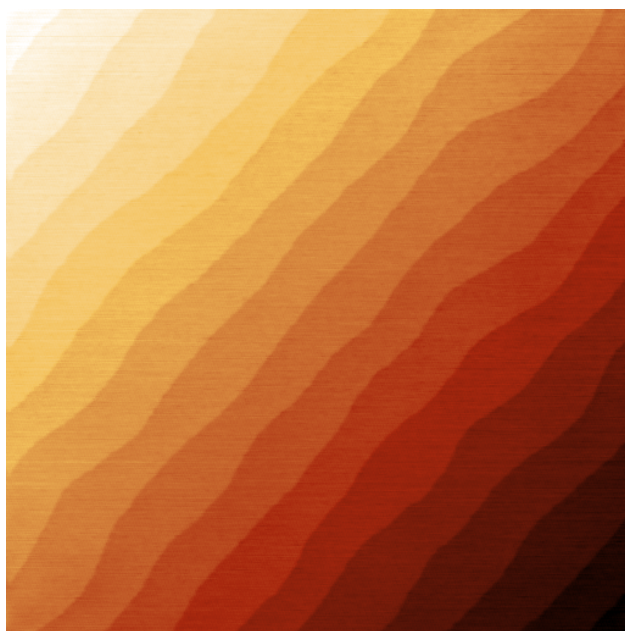


Bachelor's thesis

The influence of in-plane miscut on the surface morphology of annealed DyScO₃ (110) substrates.



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Abstract

The surface morphology of substrates¹ is of high importance for the growth of high quality thin films. For the substrate material DyScO₃ not all factors determining the surface morphology are known and large variations in surface morphology of similarly treated substrates are observed. In this report, the influence of miscut on the surface morphology of annealed DyScO₃ (110) substrates is investigated. The main question for research is what influence the in-plane miscut exercises on the surface morphology of annealed DyScO₃ (110) substrates. This study is executed by performing x-ray diffraction measurements on as received DyScO₃ (110) substrates and atomic force microscopy measurements on DyScO₃ (110) substrates after 4 hours of annealing at 1000°C. Results indicate only a marginal influence of both polar and in-plane miscut on surface morphology, and the dominant factors explaining the observed variation in surface morphology are likely not to be directly related to the miscut.

¹Cover image: Tapping Mode AFM image of DyScO₃ (110) substrate after annealing at 1000°C for 4 hours. Lateral dimension $2 \times 2 \mu\text{m}$.

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Abbreviations & symbols

DyScO ₃	Dysprosium Scandate
SrRuO ₃	Strontium Ruthenate
SrTiO ₃	Strontium Titanate
Ru	Ruthenate
SrO	Strontium Oxide
TiO ₂	Titanium Oxide
O	Oxygen
Dy	Dysprosium
Sc	Scandium
ScO ₂	Scandium Oxide
u.c.	Unit cell
e.b.s.	Electrostatic bond strength
PLD	Pulsed laser deposition
ES	Ehrlich-Schwoebel barrier
XRD	X-ray diffraction
AFM	Atomic force microscopy
TM-AFM	Tapping mode AFM
a	Lattice constant [nm]
a_{pc}	Pseudo-cubic lattice constant [nm]
θ_v	Polar miscut [°]
ϕ	in-plane miscut [°]
K_A	Overall etching constant for the AO layer [nm/min]
K_B	Overall etching constant for the BO ₂ layer [nm/min]
D	Surface diffusion coefficient [nm ² /s]
k_s	Site-to-site hopping rate [Hz]
h	Effective hopping distance between sites [nm]
V_s	Potential energy barrier from site to site [eV]
F	Deposition rate [$\mu\text{g nm}^{-2} \text{s}^{-1}$]
T	Temperature [°C] or [K]
k_B	Boltzmann constant [eV K ⁻¹]
$\kappa_{L/R}$	Probability of inclusion in left/right step edge
v	Step edge velocity [m/s]
L	Terrace length [nm]
T_c	Curie point [°C]
θ	Angle of incidence [°]
d	Spacing between parallel planes [nm]
λ	X-ray wavelength [nm]
ζ	Angle of XRD tilting stage around ζ axis [°]
ξ	Angle of XRD tilting stage around ξ axis [°]

Introduction

Perovskite type oxides like DyScO_3 and SrRuO_3 offer a wide range of interesting properties, including superconductivity, magnetism and ferroelectricity. The oxygen backbone of these materials allows the formation of highly complex strained heteroepitaxial structures and films, potentially resulting in materials with highly enhanced properties [1, 2]. DyScO_3 is a material of high interest as a substrate for heteroepitaxial structures and thin films, because of its high lattice constant and high quality crystal structure. SrRuO_3 has raised interest as a thin film material, due to unique structural, magnetic and electric properties under strain [3]. The surface morphology of the substrate is of high importance for the properties of a thin film. Surface termination, roughness and step edges all have influence on the growth of the thin film [4]. Methods using annealing and etching were previously developed in order to achieve smooth, single terminated surfaces [2, 5] for smooth thin film growth. Research has also demonstrated it to be possible to grow SrRuO_3 wires on a DyScO_3 surface using a mixed terminated surface, achieved by annealing the substrate [6].

Previous measurements [7] revealed that annealed DyScO_3 substrates from different batches sometimes show different surface morphologies after equal treatment. Particularly the level of complexity of features on step edges shows large differences for different samples with the same treatment. Highly complex step edges are expected to occur at samples with very low polar miscut angles, but do also emerge on samples with higher values of polar miscut. It is clear that not all factors influencing the surface morphology of annealed DyScO_3 (110) substrates are understood. In this report, the influence of both polar and in-plane miscut on the surface morphology of DyScO_3 substrates after annealing will be studied. The question central to this study is whether features increase in complexity, amplitude and frequency when the in-plane miscut angle increases. A maximum of feature complexity, amplitude and frequency is expected when the in-plane miscut goes towards $\pm 45^\circ$. The miscut and surface morphology of DyScO_3 (110) substrates will be studied using x-ray diffraction and atomic force microscopy.

Chapter 1 comprises the theoretical background required to study the substrate surface in relation to thin film growth. In chapter 2, an overview is given of the methods used for both measurements and analysis within this study. In chapter 3 and 4 the results from measurements and analysis are presented and discussed. Chapter 5 consists of the conclusion and several recommendations for further research. Appendix A includes a more extended presentation of several results. In appendix B, main parts of the MATLAB code developed in order to analyse the data are included.

Chapter 1

Literature review

In this chapter, a broad introduction will be given to the field of thin film growth. This will be done in the form of a review of available literature on the subjects relevant for this report. Relevant subjects are the crystal structures of relevant materials, the substrate surface and the growth of thin films.

1.1 Crystal structures

In this section, the crystal structures of the important materials for this thesis will be examined. The substrate material, Dysprosium Scandate (DyScO_3), and a thin film material, Strontium Ruthenate (SrRuO_3) will be treated. As an additional material, Strontium Titanate, SrTiO_3 , will also be treated because SrTiO_3 is well known as substrate material and more knowledge is available on SrTiO_3 compared to DyScO_3 . Both materials are in some perspectives comparable, but in order to use information on SrTiO_3 wisely for DyScO_3 , it is highly important to also know the differences.

DyScO_3 , SrTiO_3 and SrRuO_3 belong to the perovskites. Those perovskites have an ABO_3 type crystal structure. This structure contains two cations A and B and three oxygen anions. A cubic perovskite unit cell as depicted in Figure 1.1, can be characterized by alternating planes AO and BO_2 .

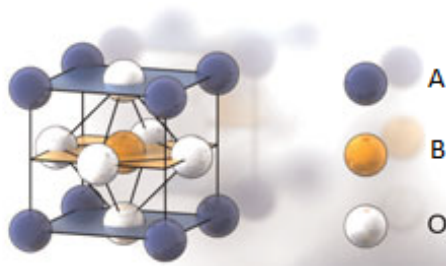


Figure 1.1: Cubic perovskite unit cell showing alternating planes of AO and BO_2 . Adapted from [6].

While SrTiO_3 has a cubic perovskite unit cell with lattice constant $a = 0.391 \text{ nm}$ [8, 9], DyScO_3 and SrRuO_3 both have a distorted orthorhombic perovskite unit cell. The distortion is the twist in the oxygen octahedra, as can be seen on Figure 1.2. On this figure, the SrRuO_3 unit cell is depicted (DyScO_3 has a similar structure). When the distortion is neglected, the orthorhombic perovskite unit cell can be represented by two pseudo-cubic unit cells on top of each other, where a pseudo-cubic unit cell is similar to the cell on Figure 1.1. In reality, the distorted orthorhombic unit cell contains 4 times the atoms of one pseudo cubic unit cell. The Ru atoms (purple on Figure 1.2) form the corners of the pseudo-cubic unit cell. From the lattice parameters of the orthorhombic unit cell, the pseudo-cubic lattice constant can be determined. DyScO_3 has orthorhombic lattice parameters $a=0.544 \text{ nm}$, $b=0.572 \text{ nm}$ and $c=0.790 \text{ nm}$ [10]. This gives a pseudo-cubic lattice constant of $a_{pc} = \frac{\sqrt{a^2+b^2}}{2} = 0.395 \text{ nm}$ [2, 6]. For SrRuO_3 , the lattice parameters are: $a=0.557 \text{ nm}$, $b=0.554 \text{ nm}$ and $c=0.785 \text{ nm}$, giving a pseudo-cubic lattice constant $a_{pc} = 0.393 \text{ nm}$ [11–13]. This means that (in the $[110]$ orthorhombic direction), both DyScO_3 and SrRuO_3 can be represented by a pseudo-cubic

crystal structure with alternating planes AO and BO_2 . With this approximation, the crystal structure of DyScO_3 looks similar to that of SrTiO_3 . An important difference however is the valence of the cations. In SrTiO_3 , Sr has a charge of 2+, where Ti has a charge of 4+. The alternating planes, consisting of either SrO or TiO_2 , are thus neutral (O has charge 2-). For DyScO_3 however, Dy and Sc both have a charge of 3+. This means that the planes are charged. The influence of this charge on the surface will be discussed at the end of subsection 1.2.1.

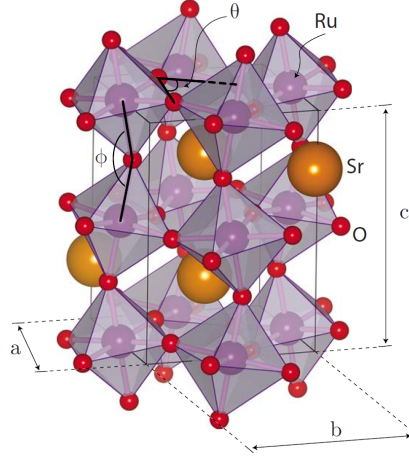


Figure 1.2: Orthorhombic perovskite unit cell of SrRuO_3 , with lattice parameters a , b and c . Adapted from [14].

Miller indices

When discussing crystals, it is sometimes necessary to describe specific points, directions or planes in the crystal lattices. The most common notation used for this is known as Miller indices [15]. These are the indices that will be (and actually already have been) used in this thesis. In order to describe a crystal structure according to the Miller indices, one first has to choose a system of coordinates. In this thesis, that will be a right-hand Cartesian system. When describing a unit cell, the origin of the system is in the left-rear-bottom corner of the cell, as can be seen in Figure 1.3. The location of each point on the unit cell is described by its x, y and z coordinates, with x, y and z normalized with the lattice parameters (thus are 1 at unit cell length). The directions are calculated by first subtracting the coordinates of two points that lie in the direction of interest. This is of course easier when one of the points is the origin. Then fractions are cleared from the result in order to give the lowest integer values. Negative integers are replaced by placing a bar over the integer and the indices are written in square brackets. This results in the following indices for $h < 0$: $[\bar{h} \ k \ l]$. Several examples of directions are shown in Figure 1.3.

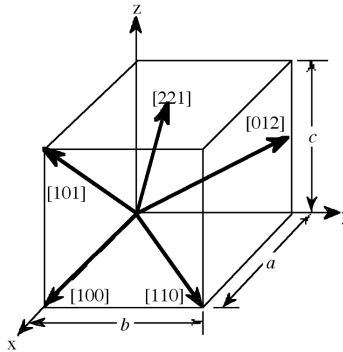


Figure 1.3: Several common directions in a cubic unit cell. Adapted from [16].

Miller indices can also be used in order to describe the multiple planes in a crystal structure. First, the coordinates at which the plane intersects with the x, y and z axes need to be identified. When the plane is parallel to an axes, ∞ is taken as the intercept. Then the reciprocals are taken of the intercepts and fractions are cleared (but the result is not reduced to the lowest integers). The resulting indices are written in parentheses, like: $(h\ k\ l)$. This procedure and several common planes can be seen in Figure 1.4.

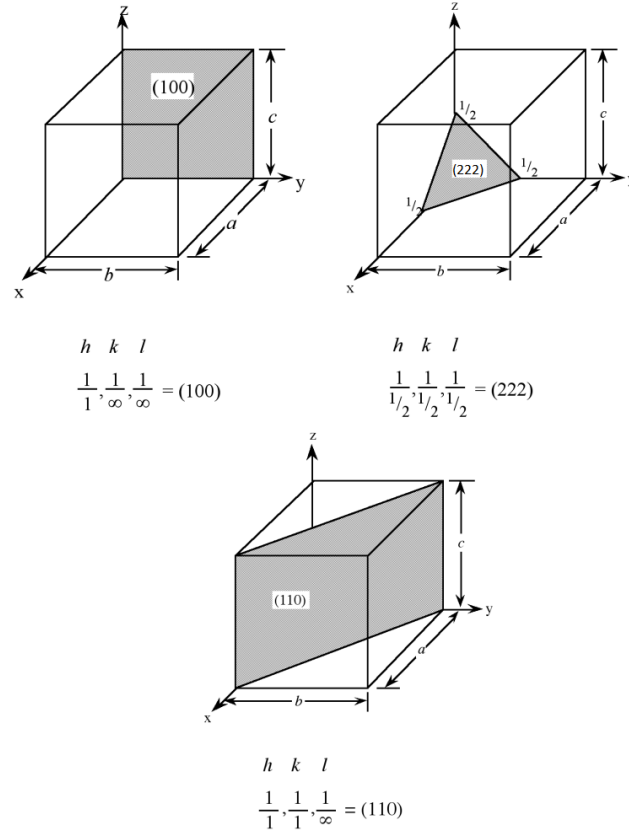


Figure 1.4: Several common planes and the calculation of their indices. Adapted from [16].

1.2 Surface

The way thin films grow, is highly dependent on the surface grown on. Therefore it is very important to understand the processes that play a role at the surface, because these processes determine the characteristics of the material. Several factors of importance are mentioned in Ref. [4]. Firstly, The sample needs to have a clean surface, free of contaminants. Secondly, Ref [17] states that the surface roughness of the substrate has to be at least of the same order as is finally required for the thin film roughness. During deposition, in many thin film materials, the surface undergoes a roughening transition. This means the initial roughness of the sample may need to be even lower. Also, the surface roughness of separate layers should not exceed the thickness of these layers. These facts together mean that only roughness on an atomic scale is allowed. Samples as received were cut and polished by the manufacturer. Though resulting in sufficiently low surface roughness, the cutting and polishing of SrTiO_3 and DyScO_3 substrates also results in a rather imperfect and mixed terminated surface, showing disordered step edges and islands on terraces with typical height differences of 0.2 and 0.4 nm (0.5 and 1 unit cell length) [2, 18], as can be seen on an AFM tapping mode image of an as received DyScO_3 (110) substrate on Figure 1.5.

For the manufacturer, it is nearly impossible to have a completely straight cut at the surface. The error when cutting the surface is referred to as the miscut. This miscut is defined with respect to the crystal

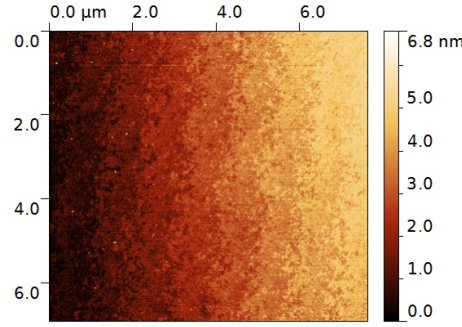


Figure 1.5: An AFM tapping mode image of a DyScO₃ substrate as received. Adapted from [7]

structure of the surface, and can be characterised by two angles, a polar angle θ_v and an in-plane angle ϕ , as is illustrated on Figure 1.6. Due to the polishing process, the miscut can effectively be different on different spots on the same substrate (both θ_v and ϕ can be different). The known influence of the miscut on the surface morphology will be discussed in subsection 1.2.2.

Although the requirements regarding roughness are fulfilled in the as received substrates, not all requirements for growth are fulfilled. As it was mentioned before, as received substrates have a rather imperfect and mixed terminated surface, showing disordered step edges and islands on terraces. For growth however, a highly ordered, highly crystalline substrate surface is necessary, because the substrate acts as a template for the desired crystalline structure.

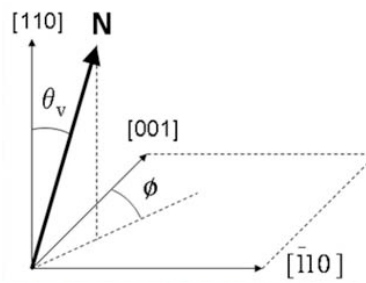


Figure 1.6: A sketch of the miscut angles - polar (θ_v) and in-plane (ϕ). $\mathbf{N}$ is the normal to the mean surface. Adapted from [19].

Another important aspect for growth is the surface termination. The (pseudo)cubic perovskite crystal structure of SrTiO₃ and DyScO₃ makes that the surface can be single or double terminated. For single termination, the surface will be completely covered with either AO or BO₂. When the surface is double (mixed) terminated, both AO and BO₂ are present at the surface. The termination of the surface is of high influence on the growth of SrRuO₃ on the surface. This influence will be elaborated further in section 1.3. For increasing the crystallinity and changing the surface termination, several treatments are developed. The main methods are annealing and etching. Both will be described in more detail in subsection 1.2.1.

As mentioned before in section 1.1, the planes in DyScO₃ are charged. In case of a perfect structure, this would mean that for single termination, the surface would be charged as well. There would be a polarity difference between bulk and vacuum. This problem is, according to Ref. [5], likely overcome by introducing oxygen vacancies in the ScO₂ surface layer or by the presence of adsorbates on the surface.

1.2.1 Treatment

In order to achieve increased crystallinity, an anneal treatment can be used. Next to this, annealing also seems to have influence on the surface termination [2, 5, 20]. In order to achieve a wider understanding of

both annealing and etching, several effects and processes observed in previous research will be mentioned and briefly described in this subsection.

Annealing

As stated before, annealing increases surface ordering and crystallinity. It does this with a process called recrystallization. During recrystallization, surface atoms diffuse and rearrange in order to decrease the surface free energy. According to Refs. [21, 22] in Ref. [18], this rearrangement leads to a regrowth of the surface forming well defined stepped terraces. This rearrangement is temperature dependent and proceeds with time. It is reported for SrTiO_3 that with increasing temperature and time steps become more and more pronounced and straight. Previous research shows that the critical temperature for this recrystallization to occur for a SrTiO_3 sample is somewhere between 700°C and 800°C . When annealing at a temperature of 650°C , no recrystallization occurs. Annealing at this low temperature however, does appear to be effective for removing residual water and organic (hydrocarbon) contamination layers from the surface [23, 24]. At temperatures higher than 1050°C and annealing time longer than 10 hours though, multiple unit cell steps and ragged stepped edges are seen [18, 23, 24].

For DyScO_3 substrates, similar results are reported. DyScO_3 as received shows steps of 0.2 and 0.4 nm (0.5 and 1 u.c.). This indicates mixed termination, for a 0.5 u.c. step means a change in termination. After annealing at 1000°C for 30 minutes, substrates show more ordering but still clear mixed termination. After 4 hours, AFM tapping mode images (see subsection 2.2.2) as on Figure 1.7a show regular and straight 0.4 nm steps [2]. This seems to indicate single termination, but Time-of-Flight Mass Spectroscopy measurements by Ref [2] show that the annealed surface still has mixed termination. This indicates that, however clearly better than before, the ordering of the surface is not entirely perfect. Ref. [6] shows that on samples annealed for 4 hours at 1000°C sometimes actually show 0.5 and 1.5 u.c. steps as can be seen in Figure 1.7b. SrRuO_3 growth on this surface (Figure 1.7c) confirms this pattern and indicates the ‘valleys’ to be ScO_2 terminated and the terraces to be DyO terminated.

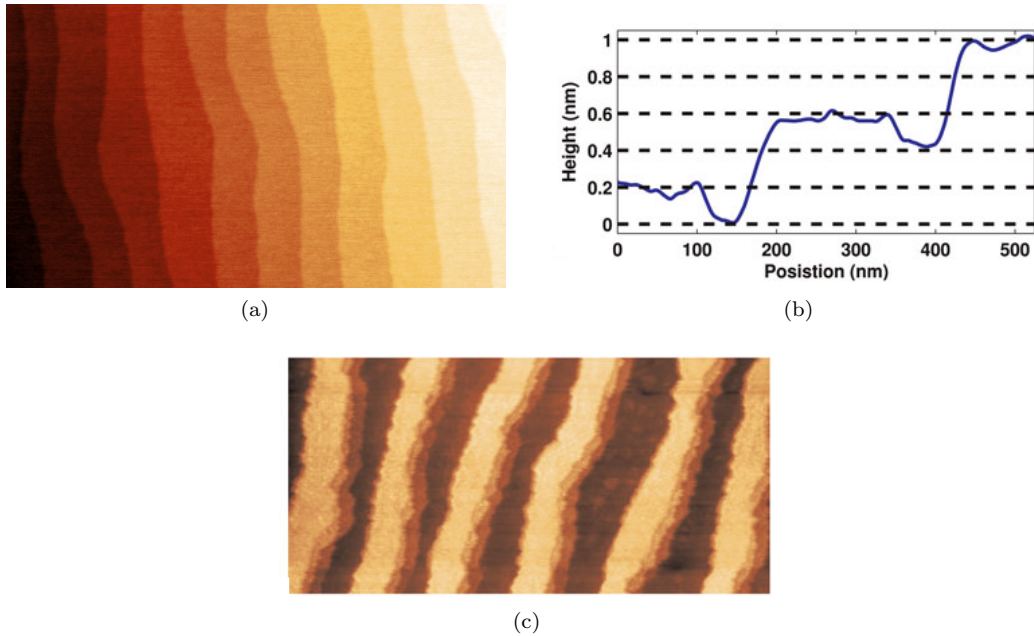


Figure 1.7: (a) TM-AFM image of 4h annealed DyScO_3 (110) at 1000°C (b): A line profile made from an AFM picture, giving detailed height information, indicating half unit-cell steps of the DyScO_3 surface after 4h annealing at 1000°C . (c): STM height map of SrRuO_3 nanowires after PLD growth on substrate with height profile (a). (a) and (b) adapted from [6].

Another variable that can be changed for annealing is the sort of environment. Ref. [4] states that annealing can either be done in an oxidising [18, 25–28] or reducing [29–34] environment. The reducing environment is often used to induce conductivity at the surface for an otherwise isolating substrate, in order to make scanning tunneling microscopy measurement possible [35, 36].

Etching

While with annealing it appears to be hard to achieve a DyScO₃ surface with complete single termination, etching is a method that makes this possible. A successful method used for achieving DyScO₃ with single termination is selective wet-etching with a basic solution like NaOH-deionized water. This method is explained in and developed by Ref. [2]. This explanation will be used below in order to form an understanding of the etching process. Selective wet-etching can be seen as a combination of two steps, firstly forming an hydroxide and secondly dissolving the hydroxide, as can be seen in Figure 1.8. This process happens for both the AO and the BO₂ layers of a perovskite.

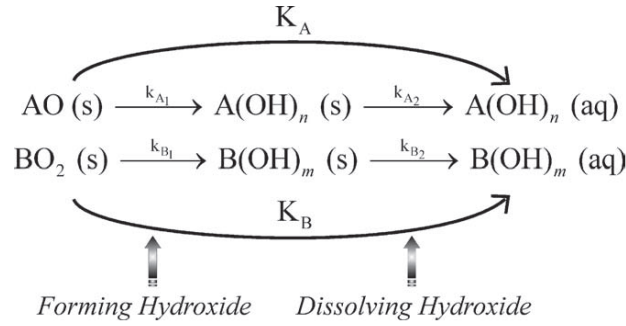
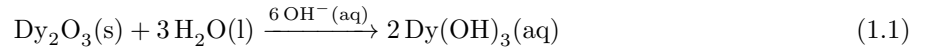


Figure 1.8: Scheme for selective wet etching of perovskite type oxides. k_{A1} , k_{A2} , k_{B1} and k_{B2} are the etching rate constants of the separate steps, and K_A and K_B are the overall etching constants for the AO and BO₂ layers. Adapted from [2].

In order to achieve single termination, the etching reaction needs to be selective towards either of both layers. In other words, the difference in the overall etching rates of AO, K_A , and BO₂, K_B , needs to be significant. When this is not the case, it is also possible to separate the steps in time, as was shown by Ref. [37] for an SrTiO₃ (001) surface. For DyScO₃ however, a selective wet-etching procedure is developed by Ref. [2]. Because both stable sesquioxides of dysprosium and scandium, Dy₂O₃ and Sc₂O₃, are evenly soluble in acidic solutions, selectivity for one of the atomic layers of DyScO₃ is expected to be minimal. In basic solutions, both sesquioxides are insoluble [38]. Modifications in the crystal structure, might induce solubility of either Sc and/or Dy in a basis etchant. In the perovskite structure, Dy and Sc have a different electrostatic bond strength (e.b.s.), due to different coordination numbers. The e.b.s. of Dy is 1/4 while that of Sc is 1/2. Due to this difference, it is expected that the following reaction dominates the etching process [2]:



The result of this should be an ScO₂ terminated surface. This is confirmed by Ref. [2]. After an anneal treatment at 1000°C of 4 hours and one hour wet-etching in 12 M NaOH–DI water, the result is an ScO₂ terminated surface with unit cell steps.

1.2.2 Miscut

One aspect that might have a large influence on the surface has not really been treated so far. This is the influence of the miscut angle on the surface morphology. As mentioned before, and as can be seen in Figure 1.6 on page 4, the miscut can be described by a polar angle θ_v and an in-plane angle ϕ . Both angles appear to have a different influence on the surface morphology after annealing. The polar angle mainly changes the ratio of terrace width and step height. An increasing polar angle, decreases the ratio of terrace width and step height. The main reason for this decreasing ratio seems to be the decreasing terrace width with increasing polar angle [5, 39].

The in-plane miscut angle seems to have a completely different influence on the surface morphology. While for DyScO_3 little is known on this subject, for (001) oriented SrTiO_3 the influence is shown in previous research. Ref. [39] has used STM imaging techniques to show the influence of the in-plane miscut on the surface morphology. The surface was cut in two ways, with zero in-plane miscut (inclined in the [100] direction), and with an in-plane miscut of 45° (inclined in the [110] direction). Both samples have a polar miscut angle of 2° . On the samples inclined in the [100] direction, straight step edges were formed after annealing (Figure 1.9a). On samples inclined in the [110] direction however, triangular edges were formed. As can be seen on Figure 1.9b, the sides of these triangular edges seem to be aligned along the [100] and [010] directions. These observations could lead to the logical conclusion that the material during annealing, after being cut, has a preference for recrystallizing along certain crystal axis.

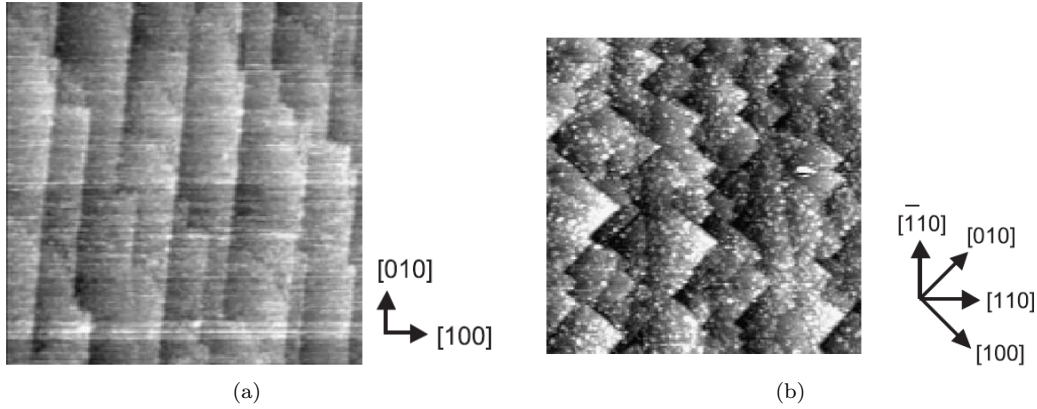


Figure 1.9: STM images of (a) a [100] inclined SrTiO_3 surface with a polar miscut angle of 2° , and (b) a [110] inclined SrTiO_3 surface with a polar miscut angle of 2° . Adapted from [39].

As mentioned before, there is little known on DyScO_3 on this subject. One could however expect the in-plane miscut angle to have influence on the surface morphology of the annealed DyScO_3 surface as well. Both the charged layers and the distorted orthorhombic perovskite crystal structure of DyScO_3 however might cause this influence to be more complex than for SrTiO_3 .

1.3 Growth

In this section, several basic principles will be explained regarding the growth of thin films. For this, several methods exist. One of those methods is pulsed laser deposition (PLD). In the following subsections, basic working principles and growth kinetics of growth by PLD will be discussed. This discussion will be specified on the growth of perovskite thin films on perovskite surfaces, SrRuO_3 on either DyScO_3 or SrTiO_3 .

Major parts of the discussion of growth kinetics, in particular the discussion about diffusion, also apply to the process of annealing. An important difference compared to depositing particles is that during annealing, particles either are already present on the surface, or arrive at the surface through bulk diffusion.

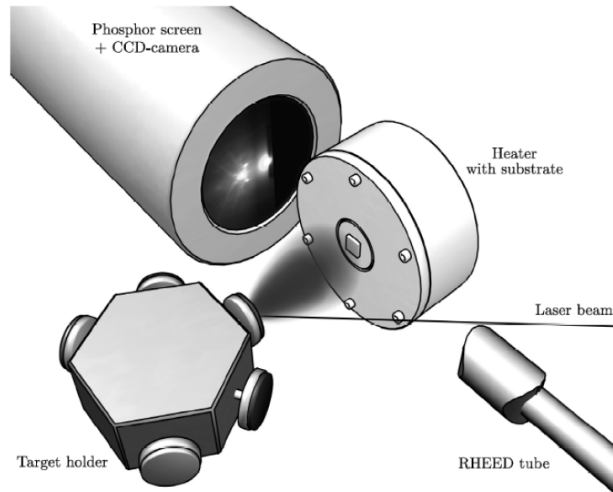


Figure 1.10: Schematic representation of a PLD with RHEED setup. Adapted from [40].

1.3.1 Basic working principles of PLD

Pulsed laser deposition is a deposition technique using (the name gives it away) a pulsed laser. A schematic representation of the technique can be seen on Figure 1.10. The laser beam is aimed at a so called target, causing the rapid removal of material from this target. This removed material forms an energetic plasma plume, which in turn condensates onto the substrate. In the process of PLD, three important phases can be distinguished: 1) the ablation and plasma formation, 2) the propagation of the plume, 3) the nucleation and growth on the substrate surface. In the ablation phase, the incoming photons are first converted into electronic excitations and then into thermal, chemical, and mechanical energy. This process results in fast removal of material from the target surface [41]. An important factor for this process is the laser wavelength. It is confirmed by Ref. [42] in Ref. [41] that this wavelength can be of high influence on the energetics of species in the plume. A very important effect of the laser wavelength is the penetration depth into the target. It is important that most of the energy of the laser is absorbed in a very thin layer near the surface of the target. This in order to prevent subsurface boiling which can lead to many particulates on the substrate film surface [41]. During the plume propagation, neutral atoms, ions and electrons travel at different velocities. Next to this, strong interactions between the species of the plasma and the background gas are observed. It sometimes seems to be important that the plume is in some degree thermalized (different species having equal lateral and forward velocities), in order to avoid resputtering of the growing film by the most energetic ions in the plume [43] in Ref. [41]. The third phase, the nucleation and growth on the substrate surface, will be extensively treated in subsection 1.3.2.

The major advantage of PLD compared to other growth techniques, is the stoichiometric removal of material from the target. Especially after an initial exposure of the target surface to the laser before deposition, the removal of material from the target preserves stoichiometry. However stoichiometric removal alone is not enough in order to ensure stoichiometric growth. It is still possible that different species incorporate into the substrate at a different rate, resputtering can occur and volatile components might evaporate from the surface. When one specie is significantly more volatile than the other species involved, then it is also possible to use a second target containing the more volatile specie. For oxide materials, it is highly important to control the oxygen content. This can be controlled by manipulating the background pressure, for PLD can be used in a wide variety of background pressures. Especially for ferroelectric materials, this adjustability makes that PLD gives the best properties with growth [41].

1.3.2 Growth kinetics

In order to understand film growth, it can be seen with different models, ranging from very simplistic to very extended. In one of the simplest models, atoms simply land on a surface, select a site and remain immobile.

If there is a certain chance for atoms to ‘stick’, and if this chance is equal for all sites, than the deposition of N particles onto N sites, leaves 37 % of the surface uncovered (while actually one monolayer of material is deposited) [41]. Obviously, real systems are more complicated than this. When the temperature is not at the absolute zero (so mostly), then there is at least some diffusion. This means that the atoms being deposited onto the surface (the adatoms), do not just arrive on the surface and select a site. The adatoms move over the surface, they diffuse. This process only stops when the atoms are immobilized, which happens when they reach an energetically favourable site. An energetically favourable site for an adatom, is a site that increases the atomic coordination of the adatom. The result of this preference for increasing their atomic coordination, is that adatoms incorporate at steps and cluster into islands when two or more adatoms meet.

The rate at which diffusion occurs, is dependent on the surface diffusion coefficient D . This coefficient is related to the site-to-site hopping rate of the adatom, k_s , by $D = h^2 k_s$, with h the effective hopping distance between sites and $k_s \propto \exp(\frac{-V_s}{k_B T})$ [44]. In this relation, V_s is the potential energy barrier from site to site, T is the temperature of the substrate and k_B is the Boltzmann constant. If the deposition rate F is constant, then the value of D determines the average distance an adatom has to travel before 1) finding an already existing island or step edge and joining it 2) meeting another adatom and nucleating to create a new island. When this process of nucleation continues, both distances decrease and eventually become constant. At this point, new adatoms being deposited will mainly join existing islands and will not form new islands any more.

An additional factor to consider is what happens when adatoms land on top of an island or terrace when deposited. At this point, it would be energetically favourable to descent of the island or terrace and be incorporated into the island or terrace edge for the resulting increase in atomic coordination. In order to descend however, the adatom needs to get pas the energetically unfavourable position on top of the step edge. This barrier it has to overcome is called the Ehrlich-Schwobel (ES) barrier [45, 46]. When this barrier is very large, adatoms will not descent of islands and terraces, with 3D growth as a result. For complex oxides, such as SrRuO_3 and SrTiO_3 however, the often observed 2D growth suggests a low ES barrier for these materials [41].

On the surfaces relevant for this thesis, step terraces will always be present. The average length of one of these terraces $L \approx \frac{a}{\theta_v}$, with θ_v the polar miscut angle (in radians) and a the (pseudo-)cubic lattice parameter of the material. When the deposition flux F is small compared to the diffusion constant D , adatoms will most likely diffuse towards the step edges instead of forming islands. There the adatoms can be included in the step edges on the left or the right, with probabilities κ_L and κ_R respectively. This step flow is illustrated in Figure 1.11. The steps S_1 and S_2 travels with a velocity of $v_1 = \kappa_L F L_2 + \kappa_R F L_1$ and $v_2 = \kappa_L F L_3 + \kappa_R F L_2$.

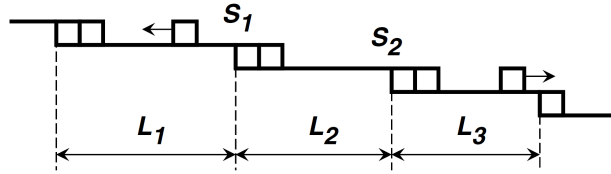


Figure 1.11: Illustration of step flow on a surface. Adatoms diffuse either to the left or to the right step edge. Adapted from [41].

The suggested low ES barrier has as a result that $\kappa_L = \kappa_R = \kappa$. If the length of the terraces is different for different steps, like $L_1 = L + \Delta L$, $L_2 = L - \Delta L$ and $L_3 = L$, this results in $\frac{v_1}{v_2} = \frac{2L}{2L - \Delta L} > 1$. This means that the larger terrace grows faster than the smaller one, which will eventually lead to step-bunching [41]. This step-bunching results in a rougher surface. While step bunching is usually attributed to this step flow, epitaxial strain also plays a part. Epitaxial strain is strain due to a lattice mismatch between crystal structure of the substrate surface and of the film. The difference in the size of the (pseudo-)cubic unit cells, causes strain in the first layers of film grown on the surface. Due to an unbalance of this strain at the step edges, step bunching can also occur [47].

An important factor for growth of thin films is also the termination of the surface. The discussion above assumes a single terminated surface. When however, as can be the case for DyScO_3 substrates, the surface

has mixed termination, this can be of influence on the growth. When the adatoms have a different diffusivity on one of the terminations, this will show during growth. SrRuO_3 has a relatively high diffusivity on DyO terminated surface areas compared to ScO_2 or on itself. This difference causes the areas with relatively low diffusivity to act as a sink for the deposited material, causing the adatoms to nucleate on these areas and to abandon the DyO terminated areas [6].

1.3.3 Strain

As mentioned before, epitaxial strain is strain due to a lattice mismatch between the crystal structure of the substrate surface and of the film. Besides the previously mentioned influence on step bunching during growth, strain also has a major influence on the physical properties of the thin film. In Ref. [1], the influence on the ferroelectric properties of the thin film are discussed. A strong polarization-strain coupling is responsible for changes in the Curie point, T_c , the paraelectric to ferroelectric transition temperature. For bulk ferroelectric ceramics and single crystals, it is long known that T_c is reduced and that piezoelectric and dielectric properties are strongly influenced by the application of hydrostatic pressure [48–50] in Ref. [1]. Thin films offer the possibility to apply (non hydrostatic) strains far above the values under which their bulk counterparts would crack. When films are sufficiently thin, biaxial strains¹ of several percent are possible without cracking the film [51–53] in Ref. [1]. Conventional believe is that the properties of any ferroelectric are optimal in a single crystal of the material. In contrast to this believe, the synthesis of highly perfect ferroelectric thin films on suitable substrates demonstrated the possibility for appropriately strained films to exhibit highly superior properties compared to single crystals. In Ref. [1], it is stated that due to the strong polarization-strain coupling, shifts of T_c of several hundreds of degrees are expected [54–63] and observed [60, 61, 64–70]. Besides influence on the ferroelectric properties, strain can also have influence on the structural and magnetic properties. For SrRuO_3 for example, previous research has shown major changes in structural and magnetic properties as a function of epitaxial strain [71–73].

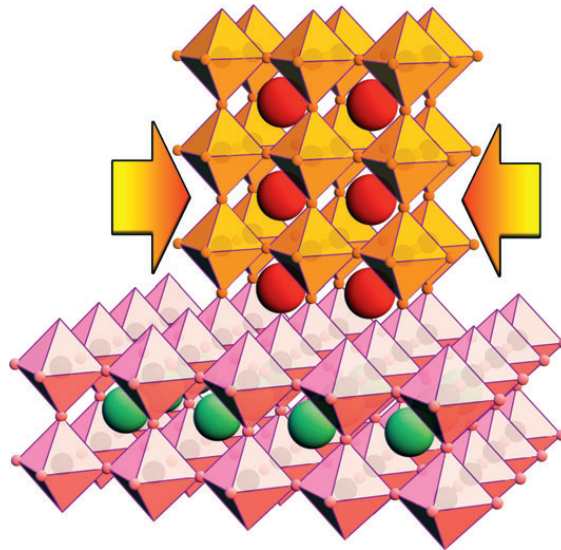


Figure 1.12: Schematic of a coherent epitaxial ferroelectric film, biaxially strained (compressive strain) in order to match the substrate. On this figure, both substrate and film have a perovskite structure. Adapted from [1].

Strain can be imposed upon thin films by a lattice mismatch or difference in thermal expansion behaviour between the substrate and the thin film, or by defects formed during film deposition [1]. The latter is undesirable because the strain field around dislocations locally alters the properties, making the ferroelectric

¹The biaxial strain is determined with the following formula, where a_0 is the lattice parameter of the strained material in its cubic state without stress and a_s is the in-plane lattice parameter of the biaxially strained material. $\epsilon_s = \frac{a_s - a_0}{a_0}$

properties of the film inhomogeneous and hereby often inferior to a coherent film. An example of a strained fully coherent epitaxial film is shown on Figure 1.12. When using a certain thin film material, like SrRuO_3 for this report, substrate materials with different lattice parameters can be used in order to impose different strains on the film. As can be seen in Figure 1.13, several commercially available substrate materials exist. A large variation of lattice parameters for the substrates is important for tuning the parameters of the thin film. SrRuO_3 has a lattice parameter in between SrTiO_3 and DyScO_3 , and different results are seen for compressive and tensile strain. About SrTiO_3 in combination to epitaxial thin film growth, a large amount of knowledge is available. On DyScO_3 , this knowledge is somewhat more limited, while the distinctive lattice parameter compared to other substrates shows potential. Therefore, this report focussing on DyScO_3 as a substrate, fits in a larger trend of research on DyScO_3 .

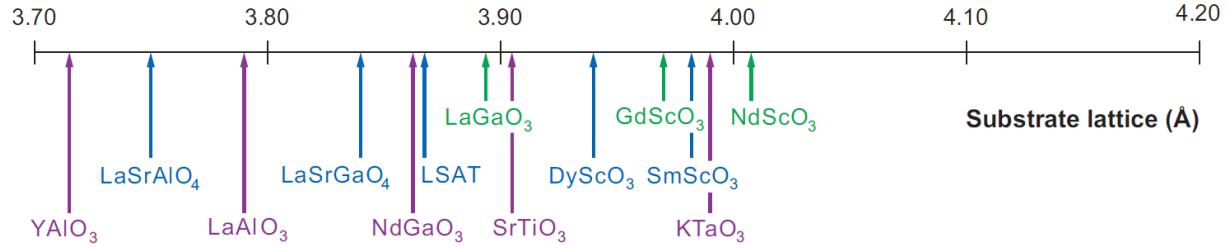


Figure 1.13: An overview of some of the perovskite or perovskite related substrates that are commercially available. The substrates are ordered by their pseudocubic lattice parameter. Adapted from [1].

Chapter 2

Methods

In this chapter, the methods used within this research will be described. First of all the sample preparation will be discussed, including the cleaning and annealing. Second the characterization of the samples by XRD and AFM will be treated. Some background information will also be given on both characterization methods. In the final part of this chapter, the methods used for analysing the data will be reviewed.

2.1 Sample selection and treatment

The DyScO₃ samples used for this research were manufactured by CrysTec GmbH using the Czochralski method. This method results in samples with a high crystallinity. As received, the samples were measured with XRD, according to the procedure discussed in subsection 2.2.1. This measurement was done in order to obtain an estimate of both polar and in-plane miscut of the samples. After these measurements 10 samples were selected for further treatment (annealing) and measurement. The samples were selected in order to get a proper idea of the influence of the in-plane miscut on the surface morphology. Samples with different in-plane miscuts were chosen in order to have sufficient variation in the miscut. In order to compare samples with the same in-plane miscut, samples were matched in pairs with similar in-plane miscut values. The influence of the polar miscut was minimized by excluding samples with extreme polar miscut values.

After this selection, the selected samples were cleaned with acetone and ethyl-ethanol, in order to rinse of any dirt left from production or from the XRD measurements. After being cleaned, the samples were annealed for 4 hours at a temperature of 1000°C in oxygen flow. In order to minimize the influence of temperature differences within the oven, or between different heating cycles, all samples used were annealed in the same cycle, on one sample holder which has a relatively small size compared to the total size of the oven (and thus minimizing eventual heat differences between samples). After this treatment, the surface morphology of the samples was checked using AFM, as will be further illustrated in subsection 2.2.2.

2.2 Characterization

In this section, the characterization techniques used within this thesis will be discussed. For XRD as well as for AFM, three aspects will be mentioned. First of all the basic working principles will be discussed, as well as different operation modes. Second, the choice for both techniques and the used modes will be explained. This second step will lead to the third step, a detailed description of the measurement procedure used.

2.2.1 X-Ray Diffraction (XRD)

XRD is an important tool used for investigating structures on the scale of atomic dimensions. When using electromagnetic radiation for investigating features with certain dimensions, the wavelength of the electromagnetic radiation used, needs to be in the same order as the features to be investigated. X-rays fit this picture for investigating structures on atomic scale, with a wavelength between 0.01 nm and 10 nm.

XRD is a relatively simple technique that can offer accurate information on interplanar spacings and atomic positions.

XRD is based on the reflection of x-rays from parallel atomic planes. For an incoming beam of x-rays, the crystal lattice appears to be a collection of atoms located in planes, as can be seen in Figure 2.1. When one parallel wave reflects from the top plane and the second reflects from the second plane, they follow a different path from the source to the detector. This difference in path length causes a phase difference between both waves when arriving at the detector. Dependent on the phase difference between both waves, constructive or destructive interference occur. Constructive interference occurs when the minima and maxima of both waves are aligned. The result is a composite wave with twice the original amplitude. When one of the waves is shifted by half a wavelength, the maxima of one wave are aligned with the minima of the other wave. Both waves will damp each other out, resulting in a composite wave with zero amplitude at all points (assuming the initial amplitudes of both waves to be the same). This is called destructive interference.

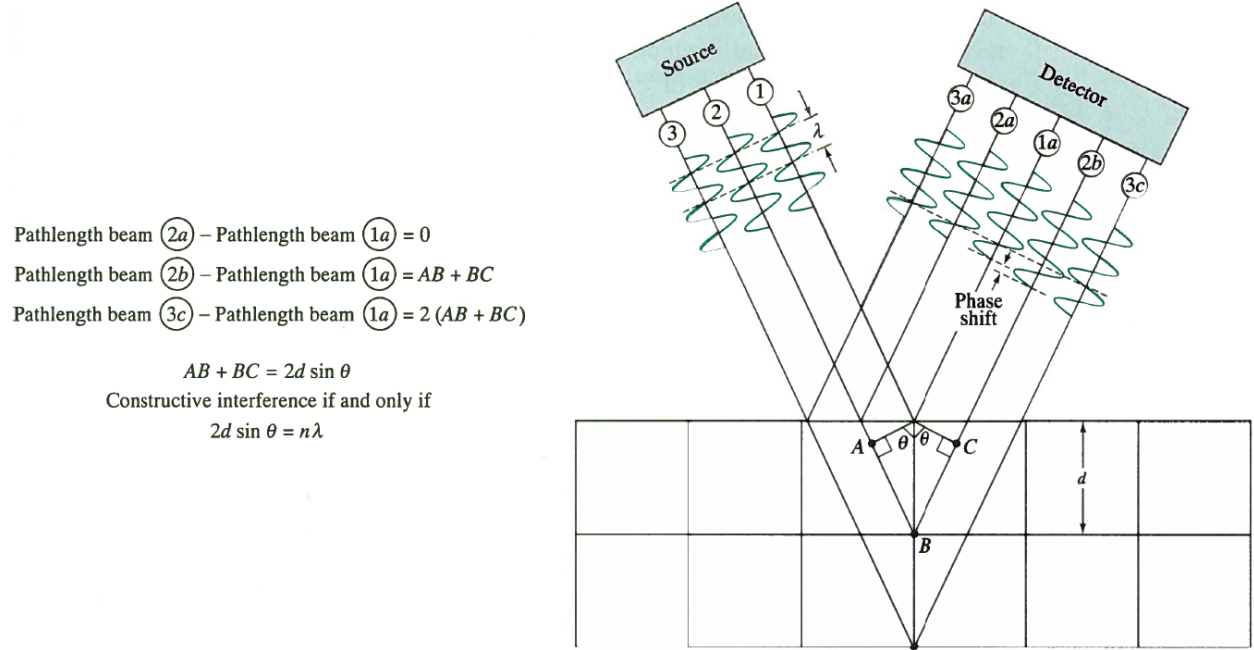


Figure 2.1: X-rays reflecting from parallel atomic planes. The incident angle is θ , the x-ray wavelength is λ , and the spacing between parallel planes is d . Adapted from [15].

The difference in path length between waves reflecting from parallel planes depends on the angle of incidence θ and the spacing between parallel planes d . When using XRD, one is searching for constructive interference. This interference is also called diffraction when occurring in an x-ray experiment. This diffraction occurs when the difference in path length equals an integer times the wavelength λ , as can be seen in Equation 2.1.

$$2d \cdot \sin \theta = n\lambda \quad (2.1)$$

This equation is well known as Bragg's law, and is central to XRD. This equation is used to extract important data from XRD measurements. It states that when the incoming wavelength and the angle of incidence are known, it is possible to determine the interplanar spacing from peaks in the spectrum (this spectrum can for example be achieved by varying the angle of incidence θ). This way information can be extracted about the interplanar spacing of a material. This information can for example be very useful for determining what material a sample consists of, to determine the interplanar spacing, or the unit cell size and lattice parameters of a specific material.

When the crystal structure of the sample is known, XRD can be used in order to retrieve information about the orientation of the sample, whether a surface is for example (110) oriented or (001), or how the

crystal lattice is oriented compared to the surface.

The latter, the difference between the surface orientation and the orientation of the crystal, is exactly what was defined in 1.2 as the miscut. This makes XRD a very suitable method to perform a first measurement of the miscut.

XRD within this report is performed on a Bruker D8 Discover, according to the following procedure. First, the sample is aligned with respect to the x-ray beam, the surface orientation is determined. This is done by letting a beam hit the sample under a very low incident angle ($\theta = 0.2^\circ$), with the detector at an equal angle compared to the sample ($2\theta = 0.4^\circ$), as can be seen in Figure 2.2a for ζ . When the surface is lying at an angle α compared to the reference for the incoming beam, then the reflecting beam from the surface will not reach the detector. The angle of the sample holder (ζ) will be optimized in order to find the angle where the detector detects the reflection. This angle is the angle where the incident angle on the surface really is 0.2° . The angle ζ also represents one of two parts of the surface orientation. The surface orientation of course has two components, so in order to determine the other component the sample is turned 90° and the procedure is repeated. This gives a second sample holder angle, ξ , which together with ζ represents the orientation of the surface.

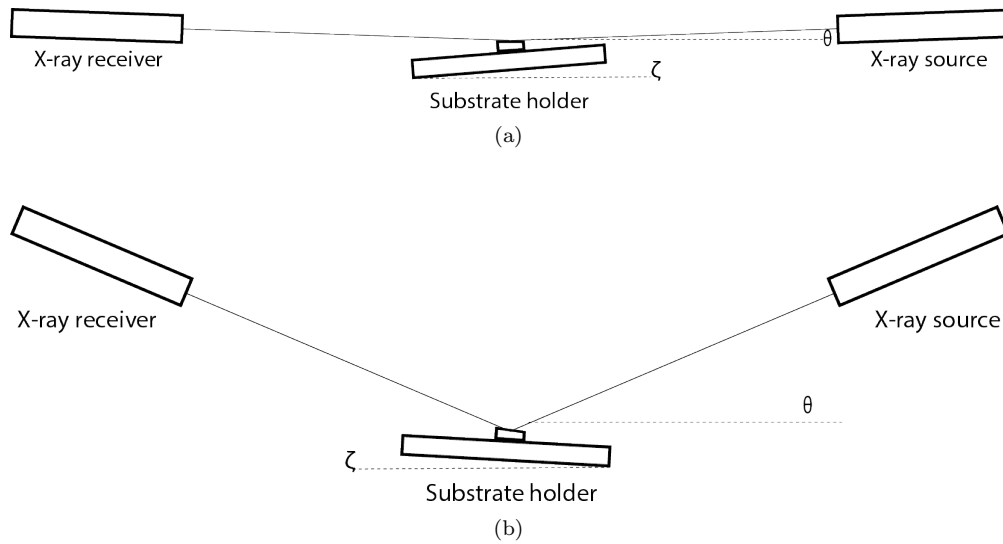


Figure 2.2: X-ray diffraction measurements. (a) Measuring the reflection from the surface with low incident angle θ and tuning ζ . (b) Measuring the diffraction from the (220) plane with incident angle $\theta = 23^\circ$ and tuning ζ .

The next step as shown on Figure 2.2b for ζ is the determination of the orientation of the crystal. The first step for determining this orientation, is to find the position of a known peak. The peak used here is the (220) peak, which is expected to be found at an incident angle of 23° . When the source and detector are properly adjusted for this peak (with a rocking curve (θ scan) and a detector (2θ) scan), ζ and ξ are determined again. This time, they represent the orientation of the crystal.

The result is two values, one for the orientation of the surface and one for the orientation of the crystal, for both ζ and ξ . The differences between both values (δ_ζ and δ_ξ) gives a representation of the miscut. In order to transform this representation of the miscut to the representation introduced in section 1.2, two equations are used. For determining the polar miscut θ_v , Equation 2.2 is used. For the in-plane miscut ϕ , Equation 2.3 is used.

$$\theta_v = \arctan \sqrt{\tan(\delta_\zeta)^2 + \tan(\delta_\xi)^2} \quad (2.2)$$

$$\phi = \arctan \frac{\tan \delta_\xi}{\tan \delta_\zeta} \quad (2.3)$$

2.2.2 Atomic Force Microscopy (AFM)

AFM, or atomic force microscopy, is a scanning technique first introduced by Gerber et al. in 1986 [74]. This technique works based on tip-sample interaction forces F_{ts} . These forces are a function of the distance between the tip and the sample, d_{ts} . In order to create an image of a surface, a sharp tip is raster scanned over the surface, and tip-sample forces are measured for every raster point. For this static mode, two scanning modes are possible. Either the z position of the tip is held constant (the tip is held on the same height) and the force F_{ts} is measured, or the force F_{ts} is fed into a feedback loop. This feedback loop is then used to keep the force F_{ts} constant while varying the height z . The first mode is generally called constant-height mode, and gives a qualitative (x, y, F_{ts}) result, where the second mode is generally called constant-force mode, and gives a quantitative (x, y, z) result [40].

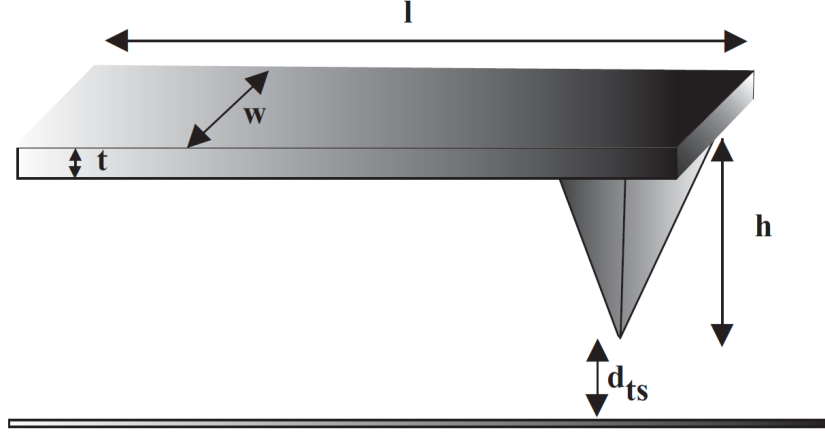


Figure 2.3: A schematic drawing of a rectangular cantilever with tip. The cantilever has length l , width w , thickness t and tip height h . The distance between tip and sample is d_{ts} . Adapted from [40].

The AFM tip is mounted on a force sensor. For this force sensor, two regular types are available, a cantilever (Figure 2.3) and a needle sensor or tuning fork. Essentially, a tuning fork consists of two coupled electromagnetic oscillators that have exactly the same eigenfrequency and oscillate in an antiparallel mode [75]. Because the tip is attached to one of the oscillators, it should either be very light in order not to change the eigenfrequency of the oscillator, or its weight should be compensated by attaching a similar weight on the other oscillator. A tuning fork is only used in dynamic mode, which will be explained below. The force sensor used within this thesis, a cantilever, can be used in both static and dynamic mode. The force F_{ts} is measured as a function of the bending of the cantilever. The cantilever can show deflection both in the z plane as in the xy plane. Measuring the latter is called friction force microscopy or lateral force microscopy. For measuring perovskite materials, different terminations can have different tip-sample interaction, leading to a contrast in lateral force. This friction mapping thus may be used for judging the termination. The cantilever has two spring constants, a torsional k_T and a normal k_N , and eigenfrequency f_0 . The bending of the cantilever can be measured using the deflection of a laser beam from the cantilever backside, as can be seen on Figure 2.4.

While the static mode has already been mentioned, AFM also has a dynamic mode. In this mode, the cantilever is oscillated such that it starts to resonate near or at its eigenfrequency. Related to the frequency are also the oscillation amplitude and phase (relative to the signal used to drive the oscillation). For dynamic mode AFM there are two main detection schemes, amplitude and frequency modulation. For amplitude modulation (AM-AFM), the cantilever is oscillating at a fixed frequency near its resonance frequency. Due to the force F_{ts} , the resonance frequency will shift, resulting in a changing amplitude at the drive frequency. This signal is fed into a feedback loop to keep the oscillation amplitude and therefore the average force in one oscillation constant. AM-AFM can be done in the attractive, intermittent and repulsive force regime. AM-AFM in the intermittent regime is often called Tapping Mode AFM (TM-AFM), and is the mode used within this thesis. This mode also allows phase imaging, registering a shift in phase between the drive signal and the measured signal. This phase shift can be inflicted by areas with different sample-tip interactions, and thus giving information similar to lateral force (friction) microscopy in static mode.

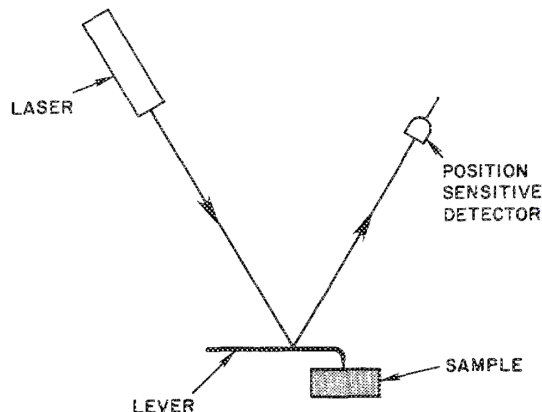


Figure 2.4: Deflection of a laser beam from the cantilever while scanning a sample. The reflection is detected with a position sensitive detector. Adapted from [76]

For this thesis, samples were measured using a Bruker Dimension Icon AFM, with Tapping-Mode tips with a thickness of $4 \pm 1 \mu\text{m}$, a length of $125 \pm 10 \mu\text{m}$, a width of $30 \pm 7.5 \mu\text{m}$, a tip height of $10 - 15 \mu\text{m}$, a force constant of $10 - 130 \text{ N/m}$ and a resonance frequency of $204 - 497 \text{ kHz}$. Because it is expected that the miscut may vary on one sample (see section 1.2), five scans are made per sample, one in the center and one close to every corner. The scansize used is $2 \mu\text{m}^2$. Information was gathered from both trace and retrace, height sensor, phase information and amplitude error.

2.3 Analysing the data

While looking at the images received with the AFM already gives a qualitative insight into the surface morphology, a more quantitative insight is desirable. In order to achieve this, information needs to be extracted regarding the surface morphology, from the images received using the AFM, especially about the level of irregularities in the step edges. A regular method for extracting information about surface morphology, is the surface roughness of the substrate. This is a measure of the texture of the surface, and can be quantified by the vertical deviations of the surface from its ideal form. When these deviations are large, the surface is rough and when they are small, the surface is smooth. On the used substrates, the smallest physically possible vertical deviations are 0.5 unit cell steps (0.198 nm). Deviations from the ideal form of the substrates on the AFM images are often observed to be smaller than this. This indicates that these deviations are measurement noise instead of true deviations of the surface. The calculation of the surface roughness would thus not give a reliable measure of the true roughness of the surface. In order to still be able to extract information about the surface morphology, several methods are developed, mainly aimed at analysing the step edges. Initial processing is performed using Gwyddion¹ and further processing is performed using MATLAB². The entire process can be divided in two stadia, preparing the data for analysis and the actual analysis.

2.3.1 Preparing the data for analysis

Preparing the data is done in four steps, illustrated in Figure 2.5 First of all, images are rotated in order to have the step edges vertical. This is necessary in order to make the images suitable for the fourth step of the preparation. The degree of rotation necessary is determined by calculating a 2D autocorrelation followed by an automatic correction of rotation in the horizontal plane. This rotation, combined with an eventual rotation angle of the scan and the angle of the sample under the AFM, results in a first measure of the in-plane miscut measured with AFM. The polar miscut is determined by counting the amount of step edges over a certain distance on an AFM image. Because every step edge stands for a 0.395 nm (1 u.c.) step, the

¹Gwyddion version 2.27

²MATLAB version R2011b

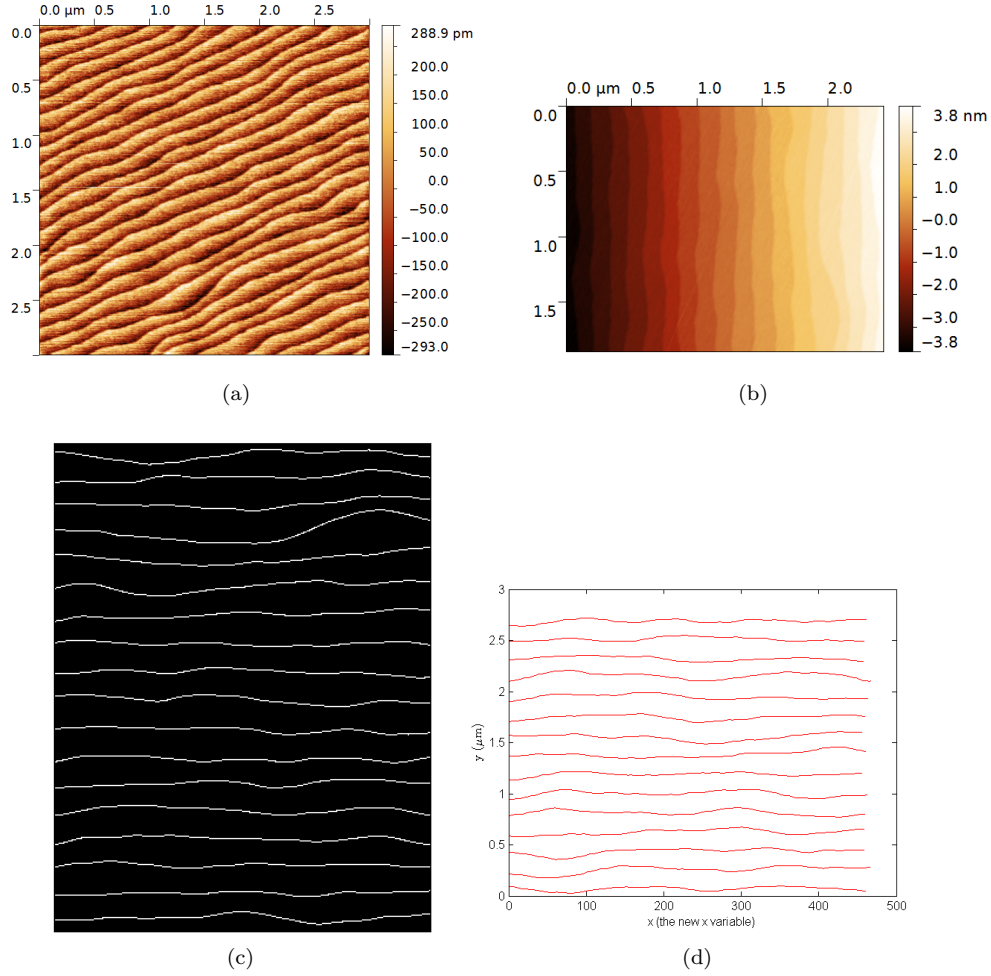


Figure 2.5: The steps preparing the data for analysis. (a) shows an original TM-AFM image, some background and scanning lines removed. (b) shows the same image rotated and levelled. (c) shows the result of the Canny method when used for this image. (d) shows the functions for the step edges extracted from (c).

polar miscut θ_v follows from Equation 2.4.

$$\theta_v = \arctan \left(\frac{(\# \text{ of steps}) \cdot \frac{0.395}{1000}}{\text{distance}(\mu\text{m})} \right) \quad (2.4)$$

A second step is to level the images in such a way that the steps become like a staircase. In the third step, the step edges are extracted from the image using the canny edge detector [77]. This method has several advantages compared to other edge detection methods (like the Sobel, Prewitt, Roberts, Lablacian of Gaussian and zero-cross method). The Canny method is partially developed for the detection of step edges on images and finds edges by looking for local maxima of the gradient of the input. This gradient is calculated using the derivative of a Gaussian filter. The Canny method is less susceptible to noise compared to other methods due to the use of two thresholds to detect strong and weak edges. Weak edges are only included in the output when connected to strong edges. The fourth step is to extract a function $y(x)$ for every step edge.

The difficulty of extracting a function from the result of the canny method is very much dependent on the presence irregularities in the step edges. The edges in Figure 2.6a can directly be extracted. The edges in Figure 2.6b however are less straightforward. When simply extracting these edges with a function $y(x)$, several x values will have multiple y values, which is of course undesirable. In order to still be able to extract

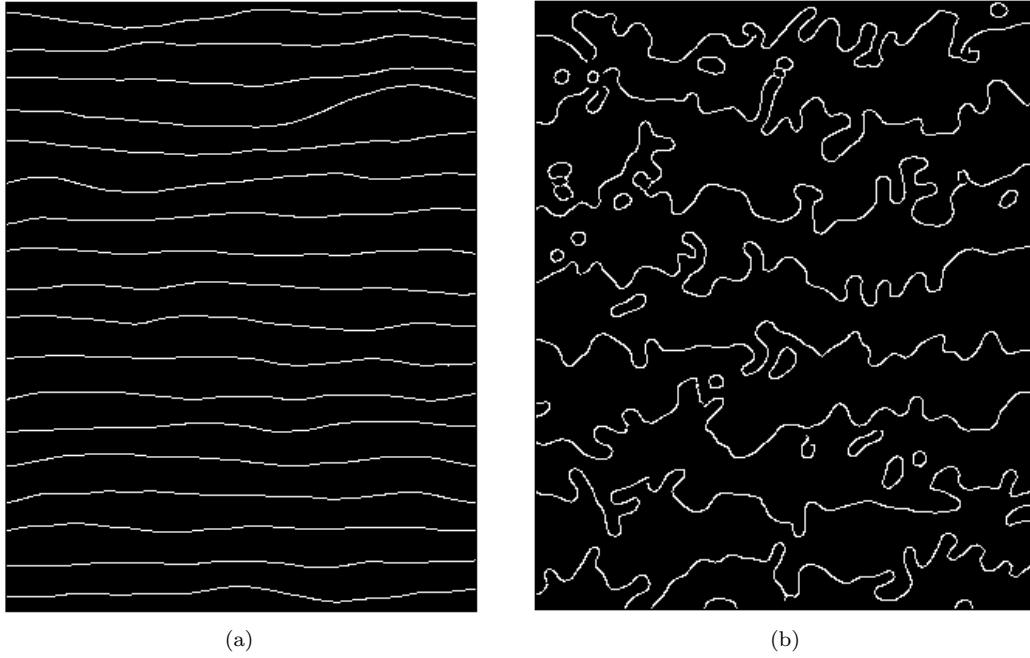


Figure 2.6: Examples of the result from the Canny method. (a): a regular profile (b): a profile with many irregularities, original TM-AFM image from Ref. [7]

the edges, a separate script was written. This script first finds the beginning of the step edges on top of the image. After that, it follows the step edges one by one, from top to bottom. At the first point, it draws a small circle around its current location and it checks whether there is a next point of the step edge somewhere on the circle edge. If not, it makes the circle a bit larger and checks again. This process continues until it has found the next point of the step edge. It saves the y coordinate of this point but discards the x coordinate, and gives it a new x coordinate based on the order wherein the script finds the point. The first point gets coordinate 1, the second 2, the third 3 etc. The advantage of this method is that every point on a step edge gets a unique x coordinate. Also the features of the step edge are preserved, which is very important for the analysis. A disadvantage is that the shape of the features sometimes changes, in order to prevent multiple y values for one x value. The results of this method for both profiles in Figure 2.6 are shown in Figure 2.7. Note that in 2.7b the features of the step edges from 2.6b have changed somewhat in shape but are still present.

2.3.2 The actual analysis

Several methods are used in order to get a quantitative insight into the step edges. These methods are enumerated here, and will be explained one by one below. While method 1 stands alone in offering a first measure of complexity of features on step edges, methods 2, 5 and 6 are aimed at measuring feature amplitude and methods 4 and 5 are aimed at measuring the frequency of features.

1. Ratio of ‘experienced steps’ perpendicular to step edges to the number of step edges
2. Standard deviation from a straight line (μm)
3. Number of crossings of a straight line along the step edges ($\frac{1}{\mu\text{m}}$)
4. Number of peaks of step edges ($\frac{1}{\mu\text{m}}$)
5. Standard deviation from a 6th and 12th degree polynomial (μm)
6. Integral power spectrum

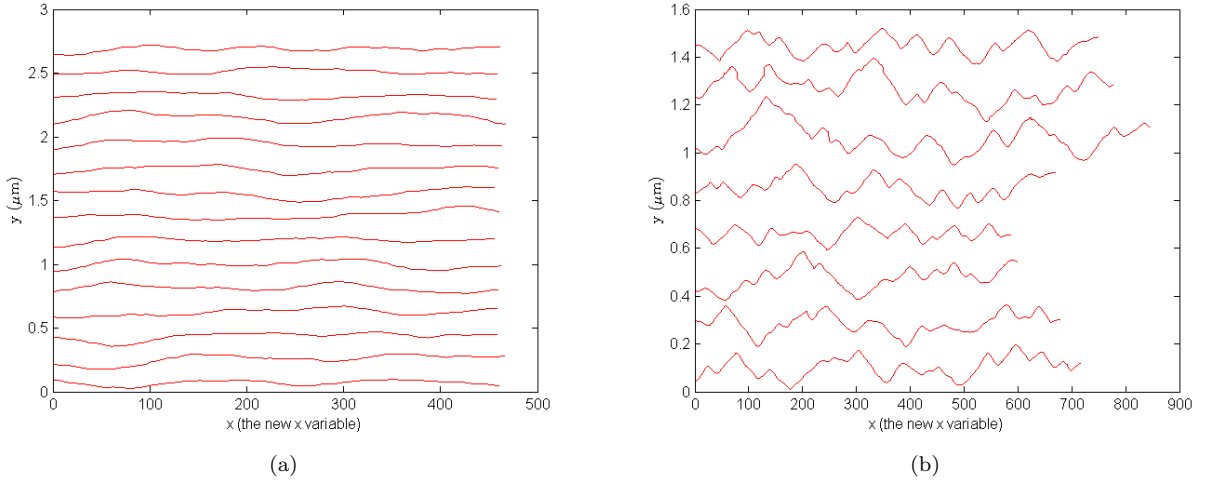


Figure 2.7: Examples of functions for the step edges as extracted from the images resulting from the Canny method on Figure 2.6. (a): a regular profile (b): a profile with many irregularities, original TM-AFM image from Ref. [7]

The first method also is the simplest method. This method takes the result from the data preparation after the canny edge method as on Figure 2.6 as input. Then it checks for every horizontal line the amount of steps it meets on the line. After this, it takes the mean value for all lines, and compares this value to the amount of steps the sample contains (this information is given to the script), giving a ratio of crossings perpendicular to step edges to the number of steps. When this ratio is larger than one, it means that step edges have features going back, or in other words, having single x -values with multiple y -values. This is a first measure of the amount of irregularities on the step edges.

The second method, the standard deviation from a straight line, takes its information from the result of the final data preparation step. For every step edge, it makes a 1^{th} degree polynomial fit of the step edge (a straight line). After subtracting this straight line from the corresponding step edge, it takes the standard deviation³ from the result. This is done for every step edge and then averaged to give an average standard deviation (in μm) for every image. This standard deviation offers information on the amplitude of the features on the step edges. The 1^{th} polynomial is also used to correct the measurement of in-plane miscut mentioned in subsection 2.3.1. For every step edge, the angle this polynomial (straight line) has compared to a line with zero slope. The values for every step are averaged to give a value for the entire sample, which is used to correct the measure of in-plane miscut mentioned in subsection 2.2.2. The resulting measure of in-plane miscut will be used for the analysis.

The third method uses the 1^{th} degree polynomial fit from the second method. It lays this line over the step edge and looks for places where this line crosses the step edge. The more features the step edge possesses, the more the line crosses the step edge. The script repeats this for every step edge and calculates the mean number of crossings per μm for the entire sample.

The fourth method uses the representation of the step edges as on Figure 2.5d on page 18. For every step edge, it then calculates the number of peaks and averages this over all step edges of the sample. The result is the average amount of peaks per μm for a sample.

The fifth method uses a 6^{th} as well as a 12^{th} degree polynomial fit for approximating the step edges. After the fit, it calculates the quality of the fit. It does this by calculating the standard deviation of the current step edge from the fit. The information this method provides becomes relevant when compared to the standard deviation from a straight line (the second method). The standard deviation from the straight line (1^{th} degree) compared to the deviation from the 6^{th} and 12^{th} degree provide information about the amplitude

³Standard deviation: $\sigma = \sqrt{\frac{1}{n} \sum_{i=1}^n (x_i - \bar{x})^2}$

and frequency of the features. While not executed within this report, this method offers the possibility to make a more extended analysis by analysing the polynomial coefficients. These coefficients offer information about the presence of higher order features. For this expansion, a higher order polynomial fit may be used in order to get the most comprehensive description possible.

The sixth method makes use of a Fourier transform. For every separate step edge a Fourier transform is performed, after a 1th degree polynomial fit is subtracted from the edge in order to cancel the effect of the angle under which the edge is lying. The result is then converted into a power spectrum, according to Equation 2.5. This power spectrum gives a measure for the total information contained at a certain frequency. The power spectrum is then integrated, in order to yield the total surface under the power spectrum. This total surface offers information about the deviation of the step edges from a straight line. This method offers the possibility for expansion in terms of a more extended evaluation of different frequency domains. This may offer information about the presence of certain frequencies in the profile of the step edges.

$$\text{Power} = |\text{freq}|^2 \tag{2.5}$$

Chapter 3

Experimental results

In this chapter, the results from the experiments and the analysis will be presented. First, intermediate results from the XRD and AFM measurements will be presented. Thereafter the results from the previously mentioned methods of analysis will be discussed.

3.1 Results XRD and AFM measurements

Both XRD and AFM measurements were used in order to determine the miscut for the ten selected samples. While XRD measurements were done ones per sample, multiple images per sample (five) were made with AFM. The results of these measurements are summarized in Table 3.1 for the polar miscut θ_v , and in Table 3.2 on the following page for the in-plane miscut ϕ . A more complete overview of the results is shown on Table A.1

Sample	θ_v XRD ($^\circ$)	Avg θ_v AFM ($^\circ$)	Std deviation ($^\circ$)	Difference with XRD ($^\circ$)	absolute values ($^\circ$)
DSO03	0.150	0.167	0.022	0.017	0.017
DSO04	0.190	0.181	0.021	-0.009	0.009
DSO05	0.160	0.171	0.048	0.011	0.011
DSO08	0.180	0.177	0.029	-0.003	0.003
DSO11	0.160	0.179	0.026	0.019	0.019
DSO12	0.200	0.181	0.021	-0.019	0.019
DSO13	0.160	0.168	0.018	0.008	0.008
DSO14	0.090	0.153	0.019	0.063	0.063
DSO17	0.130	0.148	0.012	0.018	0.018
DSO19	0.120	0.121	0.024	0.001	0.001
Average	0.154	0.165	0.024	0.011	0.017

Table 3.1: Further comparison of polar miscut measurements between XRD and AFM. The Avg θ_v AFM is the average value of the polar miscuts of the different locations on one sample. The Std deviation is the standard deviation of the polar miscuts on the different locations of one sample from the average value. The difference with XRD is the difference between the average AFM value and the XRD value, where the absolute value is the absolute value of the latter.

and Table A.2 in Appendix A. The large difference between the values measured with XRD and AFM are striking when examining both tables. Also striking is the large variation of values measured with AFM for θ_v and ϕ within one sample. This is also expressed by the relatively large standard deviations within one sample for both θ_v and ϕ , as well as the relatively large differences between XRD and AFM measurement. It is also striking that the standard deviation of the XRD measurements from the average values of the AFM measurements, is in the same order as the average standard deviation of the AFM measurements. This shows for both the θ_v as the ϕ measurements, as for θ_v the value of the standard deviation of the XRD

Sample	ϕ XRD ($^\circ$)	avg ϕ AFM ($^\circ$)	Std ($^\circ$)	Difference with XRD ($^\circ$)	absolute values ($^\circ$)
DSO03	34.66	32.23	6.70	-2.43	2.43
DSO04	69.41	64.43	4.75	-4.98	4.98
DSO05	68.44	61.36	14.51	-7.08	7.08
DSO08	36.07	42.15	7.07	6.08	6.08
DSO11	-54.69	-59.15	7.11	-4.46	4.46
DSO12	-53.62	-54.50	6.57	-0.88	0.88
DSO13	47.01	61.07	5.71	14.06	14.06
DSO14	-7.69	-8.15	8.66	-0.46	0.46
DSO17	0.59	-5.22	10.03	-5.81	5.81
DSO19	-44.55	-50.02	7.96	-5.47	5.47
Average	9.563	8.42	7.91	-1.14	5.17

Table 3.2: Further comparison of in-plane miscut measurements between XRD and AFM. The Avg ϕ is the average value of the in-plane miscuts of the different locations on one sample. The Std deviation is the standard deviation of the in-plane miscuts on the different locations of one sample from the average value. The difference with XRD is the difference between the average AFM value and the XRD value, where the absolute value is the absolute value of the latter.

measurements from the average values of the AFM measurements is 0.025° and for ϕ this value is 6.67° . Note that this is indeed in the same order as the average standard deviations within the AFM images of 0.024° for θ a somewhat larger value for ϕ of 7.91° .

3.1.1 Errors in measurement

Both measurements by XRD and AFM only use part of the surface. With AFM 5 scans of $2\mu\text{m}^2$ are made at different spots for every sample. Only a relatively small part of the surface has thus been used in order to determine the miscut. With XRD, one scan is made for every sample. The x-ray source emits a line of x-rays. This line has a footprint that is higher than the used samples, and thus completely covers the samples in one direction. The width of the line is smaller than the width of the samples. The measurement is an average of an area on the sample as high as the sample and as wide as the x-ray line where it hits the sample. The measurement by XRD is based on a larger area compared to the measurement by AFM.

A possible cause for error in the XRD measurement, is the fact that the sample is rotated 90° between measuring ζ and ξ . This results in both angles being measured at a different part of the surface, with only a small overlap. Due to the large variation in the miscut on different spots on the same sample as shown with the AFM images, this difference can cause an error in the XRD miscut measurement.

With AFM, the shifting of the image during scanning can cause an error in the determination of the in-plane miscut by AFM. This shifting means that what appears to be a curved step edge on an AFM image, could be a straight edge in reality. This curvature could cause an error while determining the in-plane miscut from the AFM images.

3.2 Results from analysis

In this section, the results from the previously mentioned methods of analysis will be discussed. This discussion will be conducted in two steps. First, the results from the different AFM images will be combined to give results for the different samples. In the second step, results from individual images will be compared based on the miscut, disregarding the sample images originate from.

3.2.1 Comparison of complete samples

The comparison of complete samples will be done using both the original pairs defined based on XRD measurements of miscut and a new ordering of samples based on the AFM measurements of the in-plane

miscut. In the new ordering both sample DSO03 and DSO08 are not included in any set any more and DSO04, DSO05, DSO11 and DSO13 form a set of 4 samples. The results as well as both the original and new pairs for the complete samples are shown in Table 3.3 and Table 3.4. In Figure 3.1, multiple plots are shown of the normalized results of the various methods plotted against the in-plane miscut measured by AFM. Results were plotted against the average miscut measured by AFM, because it is assumed that the five AFM measurements provide a better approximation of the in-plane miscut for the whole sample compared to one XRD measurement.

Figure 3.1a shows the results of the method using the ratio of ‘experienced steps’ perpendicular to step edges to the number of step edges. Figure 3.1c shows the result of the methods aimed at measuring the amplitude of features and Figure 3.1e shows the result of the methods aimed at measuring the frequency of features. Note that the different methods aimed at measuring the amplitude show relatively similar trends, as well as the methods aimed at measuring frequency. A connection between the in-plane miscut and amplitude, frequency or ‘ratio’ is not apparent from Figure 3.1a,c,e. Note from Table 3.3, that the two sample pairs (DSO04-DSO05 and DSO14-DSO17) with the respectively the highest and lowest miscut (measured by XRD), have relatively low values for both the # of crossings and the # of peaks. For the other measures in Table 3.3 and Table 3.4, no connection is apparent between miscut and results.

When the AFM images are examined without further measures of analysis, it appears that none of the samples used shows features as shown on Figure 2.6b on page 19. In order to demonstrate the effect on the results from the methods of analysis, three samples (TM-AFM images shown in Figure 3.2) showing such features are added to the data. These samples have had similar treatment (cleaning with ethanol, 4h annealing at 1000°C) as the samples in the original dataset. Miscuts and results for these samples are shown in Table 3.5. Important note is that for the added samples, only one AFM figure was made per sample, and no XRD data is available. With the determination of in-plane miscut, a larger error is expected for the exact angle of the samples in the AFM is not known.

A graphical representation of the results for the extended dataset is shown in Figure 3.1b,d,f. Note the three in-plane miscut values where peak values occur. These values represent the added samples. Also note that the first in-plane miscut value with peak values (representing DSO21) gives particularly high values for the methods measuring amplitude, while the number of crossings and peaks per micrometer is actually lower than average. Further, note that most of the variation present in Figure 3.1a,c,e largely disappears compared to the new peaks in Figure 3.1b,d,f.

Sample	original pairs	new pairs	ϕ XRD ($^{\circ}$)	ϕ AFM ($^{\circ}$)	Ratio (1)	std. dev. (μm) (2)	# of crossings ($\frac{1}{\mu\text{m}}$) (3)	# of peaks ($\frac{1}{\mu\text{m}}$) (4)
DSO03	1		34.66	32.23	0.998	0.016	3.76	3.23
DSO04	2	1	69.41	64.43	0.999	0.015	3.55	2.64
DSO05	2	1	68.44	61.36	1.000	0.018	3.06	1.68
DSO08	1		36.07	42.15	1.000	0.014	3.61	2.65
DSO11	3	1	-54.69	-59.15	0.999	0.015	4.07	2.57
DSO12	3	2	-53.62	-54.50	0.999	0.012	4.03	3.15
DSO13	4	1	47.01	61.07	1.000	0.017	4.11	3.32
DSO14	5	3	-7.69	-8.15	0.999	0.020	2.76	2.09
DSO17	5	3	0.59	-5.22	1.000	0.022	3.12	2.55
DSO19	4	2	-44.55	-50.02	1.002	0.021	4.02	2.98

Table 3.3: Results of analysis for complete samples part 1. The (#) stands for the number of the method in the enumeration in Subsection 2.3.2 on page 19.

Sample	original pairs	new pairs	ϕ XRD ($^{\circ}$)	ϕ AFM ($^{\circ}$)	std. dev. 6th deg. (μm) (5)	std. dev. 12th deg. (μm) (5)	Integral power spectrum (6)
DSO03	1		34.66	32.23	1.50	0.061	1.58
DSO04	2	1	69.41	64.43	1.48	0.052	1.88
DSO05	2	1	68.44	61.36	1.41	0.035	1.34
DSO08	1		36.07	42.15	1.12	0.044	1.34
DSO11	3	1	-54.69	-59.15	1.43	0.049	1.50
DSO12	3	2	-53.62	-54.50	1.12	0.046	0.99
DSO13	4	1	47.01	61.07	1.41	0.057	1.75
DSO14	5	3	-7.69	-8.15	1.39	0.063	2.28
DSO17	5	3	0.59	-5.22	1.93	0.085	3.13
DSO19	4	2	-44.55	-50.02	1.95	0.090	2.01

Table 3.4: Results of analysis for complete samples part 2. The (#) stands for the number of the method in the enumeration in Subsection 2.3.2 on page 19.

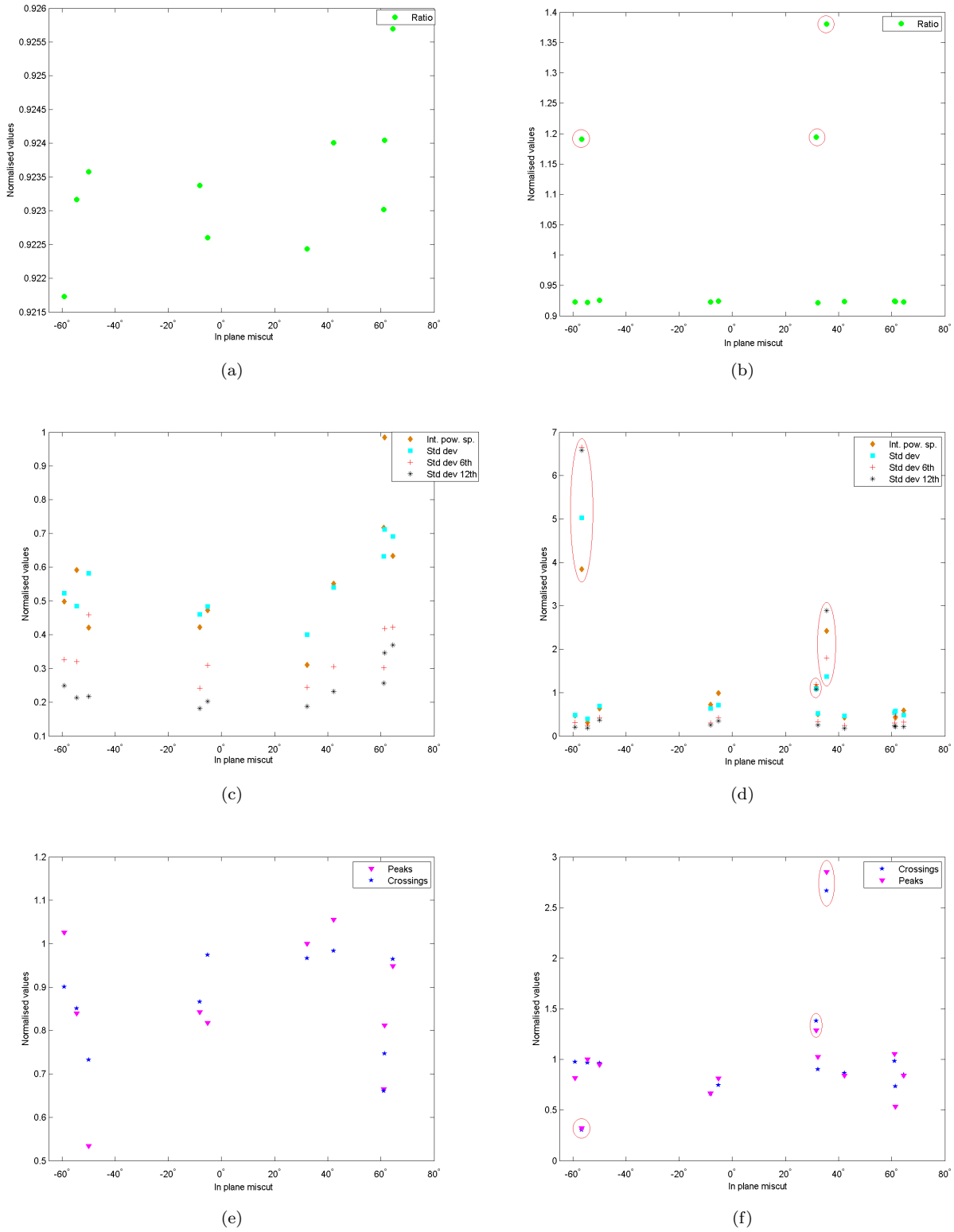


Figure 3.1: Normalised results of the several methods of analysis plotted against the in-plane miscut measured by AFM. Results averaged per sample. Figures a,c,e show results for the original dataset, for respectively the ‘ratio’ method, methods aimed at feature amplitude and methods aimed at feature frequency. Figures b,d,f show results for the extended dataset (results added samples encircled in red), for respectively the ‘ratio’ method, methods aimed at feature amplitude and methods aimed at feature frequency.

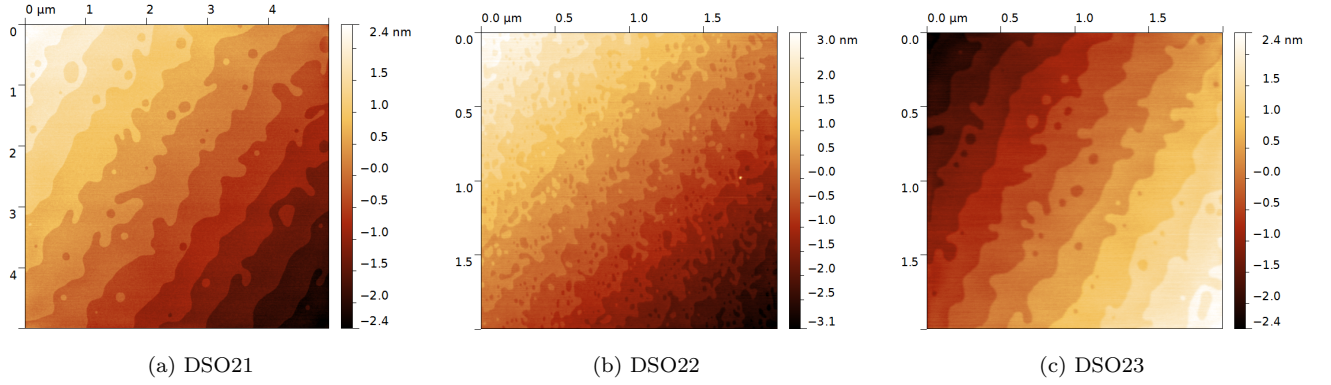


Figure 3.2: TM-AFM figures of samples with more complex features. Figure for DSO21 from Ref. [78], figures for DSO22 and DSO23 from Ref. [7].

Sample	θ_v XRD ($^\circ$)	ϕ AFM ($^\circ$)	Ratio (1)	std. dev. (μm) (2)	# of crossings ($\frac{1}{\mu\text{m}}$) (3)	# of peaks ($\frac{1}{\mu\text{m}}$) (4)
DSO21	0.041	-56.64	1.29	0.155	1.27	1.02
DSO22	0.125	35.49	1.49	0.042	11.13	8.96
DSO23	0.096	31.65	1.29	0.034	5.76	4.05

Sample	θ_v XRD ($^\circ$)	ϕ AFM ($^\circ$)	std. dev. 6th deg. (μm) (5)	std. dev. 12th deg. (μm) (5)	Integral power spectrum (6)
DSO21	0.041	-56.64	0.060	0.0031	12.22
DSO22	0.125	35.49	0.032	0.0028	7.67
DSO23	0.096	31.65	0.021	0.0010	3.59

Table 3.5: Parameters and results for samples added to the analysis. The (#) stands for the number of the method in the enumeration in Subsection 2.3.2 on page 19.

3.2.2 Results from individual images

In this subsection, results from individual images will be compared. This analysis includes the extended dataset (the ten selected samples as well as the three added samples). In Figure 3.3a,c,e, the results of the different methods are plotted against the in-plane miscut. Note again the presence of three major peak values (two close together around 35°) for the measures of feature amplitude and the ‘ratio’ method. Also note that for original dataset, no clear trend can be distinguished. In Figure 3.3b,d,f, the results of the different methods are plotted against the polar miscut. On Figure 3.3d, a slightly descending trend can be recognised, indicating a connection between polar miscut and feature amplitude.

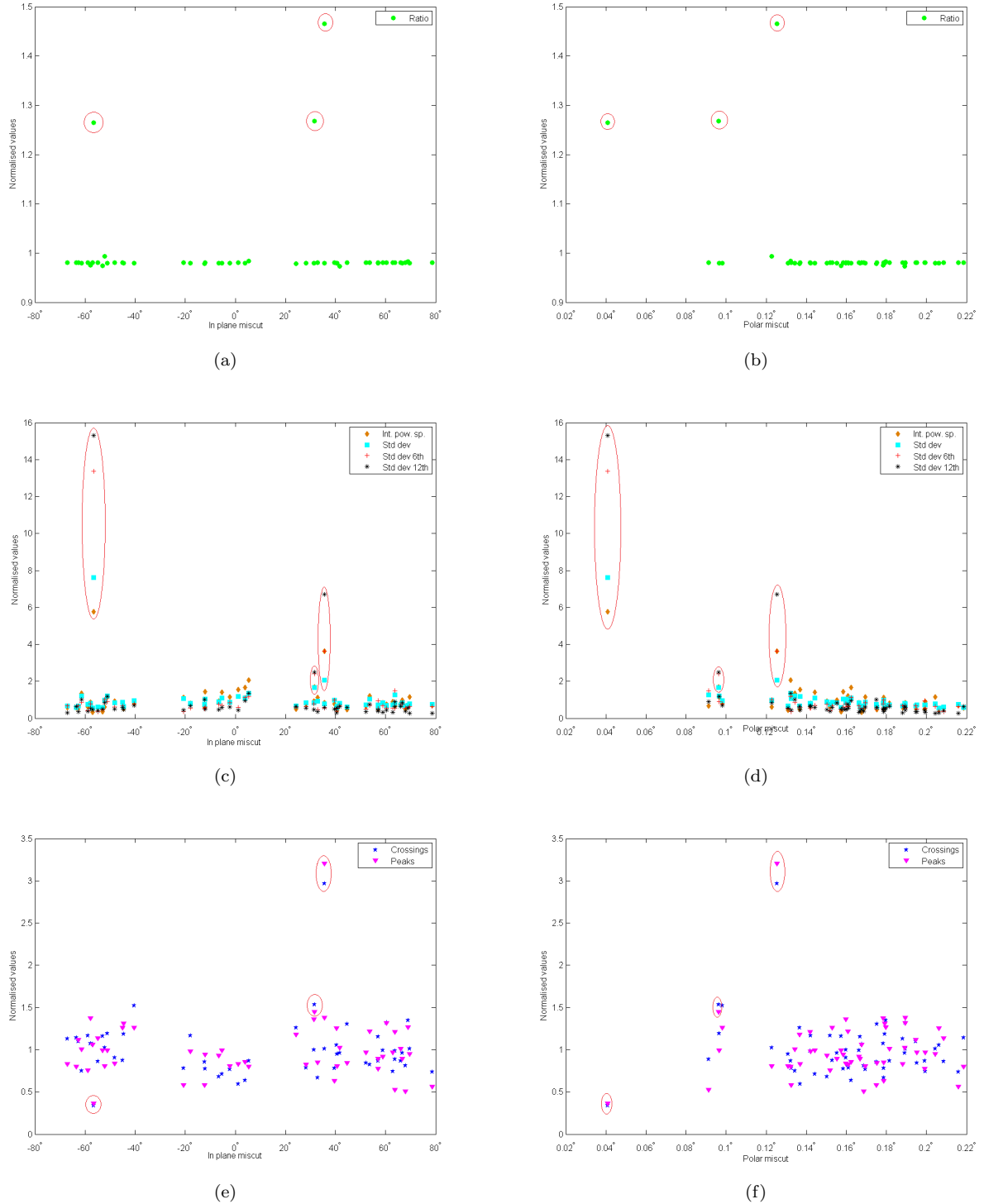


Figure 3.3: Normalised results of the several methods of analysis for individual images from the extended dataset (results added samples encircled in red). Figures a,c,e show results of respectively the ‘ratio’ method, methods aimed at feature amplitude and methods aimed at feature frequency plotted against the in-plane miscut. Figures b,d,f show results of respectively the ‘ratio’ method, methods aimed at feature amplitude and methods aimed at feature frequency plotted against the polar miscut.

Chapter 4

Discussion

In this chapter, the implications of the results as presented in chapter 3 will be discussed. Firstly, the intermediate results from the XRD and AFM measurements will be interpreted. Secondly, the results from the developed methods of analysis will be interpreted. Finally, the performance of methods of analysis will be discussed.

4.1 Results XRD and AFM measurements

As mentioned before in section 3.1, the value of the standard deviation of the XRD measurements from the average value of the AFM measurements is 0.025° for θ_v and 6.67° for ϕ . The average standard deviations within the AFM images are 0.024° for θ_v and 7.91° for ϕ . Measurements by XRD thus have a standard deviation in a similar order of size as AFM measurements, though the standard deviation for θ_v measured with XRD is somewhat smaller than the standard deviation for θ_v measured with AFM. A possible explanation for these results can be found in the measurement errors mentioned in subsection 3.1.1. Based on the size of the XRD measurement compared to the AFM measurement, one would expect XRD to give a better representation of the average value than a $2\mu\text{m}^2$ AFM image of the surface. The error caused by rotating the sample between measuring ζ and ξ , might however result in a larger error in the XRD measurement, causing the standard deviation to be similar to the measurement by AFM. The larger error in the AFM measurement of in-plane miscut could be caused by the shifting of the image during the scanning process. This shifting, as mentioned in subsection 3.1.1, could mean that what appears to be a curved step edge on an AFM image, could be a straight edge in reality. This curvature could cause an error while determining the in-plane miscut from the AFM images.

4.2 Results from analysis

Samples with both the lowest and the highest miscut, yield relatively low values for the number of crossings of a straight line along the step edges and the number of peaks per step edge. This could be an understandable indication for a connection between the in-plane miscut and the features, because both an in-plane miscut of 0° and of 90° would mean that the sample is cut along a crystal axis. No clear connection between in-plane miscut and features on the step edges is found with methods focused on amplitude of the features or the ‘ratio’ method.

Despite the fact that the samples were selected on a limited range of polar miscut values, still the results seem to reveal a connection between the polar miscut and the amplitude of features of the step edges. It appears that, with an increasing polar miscut, the amplitude of features measured by the analysis is reduced. This could be explained by the coupling of the polar miscut and the terrace width. A larger polar miscut means a smaller terrace width (subsection 1.2.2), and thus a smaller distance for atoms to diffuse during annealing in order to be integrated into a step edge. A smaller angle means a larger distance for atoms to travel before being integrated into a step edge. This may cause atoms to cluster, eventually causing step edges with larger irregularities. The marginality of the connection could be explained by the limited range of

values for the polar miscut in the dataset. Also note that the results of analysis methods aimed on feature amplitude report the highest amplitude for the sample with the lowest polar miscut, DSO21. The methods aimed at measuring the frequency, yield relatively low frequencies for this sample. Both methods even yield a frequency for this sample lower than the average for all samples. This can also be explained with the low polar miscut of this sample, for larger features also mean a low spatial frequency of features. In other words, the number of crossings per μm as well as the amount of peaks per μm is relatively low on samples with a low polar miscut.

The samples added in the extended dataset, show significantly different results from the samples in the original dataset, especially for the ‘ratio’ method and methods aimed at feature amplitude. This difference indicates the influence of more dominant factors on features than the miscut, for the variation of miscut does not explain those features.

4.3 Performance methods of analysis

In this section, the individual and combined performance of the different methods of analysis will be discussed. Both strengths and weaknesses of the methods will be reviewed.

The ‘ratio’ method provides a very basic and reliable detection of the presence of features when these are causing the functions for step edges to have single x-values with multiple y-values (features ‘going back’). It will only give a ‘result’ when these complex features are present. The methods using the standard deviation from a straight line, from a 6th and 12th degree polynomial, and the method using a fourier transform provide information about the amplitude of the features present. Comparing these methods shows that the results from the different methods are very similar. This indicates that these methods do all measure the same variable, and form a plausible measure of feature amplitude. Also these methods are all very well able to distinguish the ‘added samples’ (with complex features) from the ‘original samples’ (without complex features). However the added value of more than one measure is limited. In order to add value to the analysis, both the method using a 6th and 12th degree polynomial and the method using a fourier transform can be expanded. Possible ways to expand these methods are discussed in chapter 5. A present weakness of the method using the fourier transform, is that the result from this method is also dependant on the length of a step edge. This means that when a larger image is reviewed, this method will give a different result. This difference is caused by the fact that when a step edge becomes longer, also more frequencies will be present (or more frequencies are necessary to approximate the step edge). In order to properly use this method, the result needs to be corrected for the length of the step edges on an image.

The remaining two methods, calculating the number of crossings in a straight line along the step edges and calculating the amount of peaks in the step edges, provide information about the average frequency of features. Both methods give somewhat different results compared to the previously mentioned methods, but compared to each other results are very similar. The difference with other measures is as expected, for these methods measure the frequency and not the amplitude of features. Samples with obvious high frequencies are also recognised by both methods. These methods also offer the possibility to distinguish between samples with complex features as seen on samples with a relatively low miscut, and with complex features as seen on samples with more regular miscuts. A weakness of the method calculating the number of crossings, is the straight line it uses. Due to features of different size, the straight line means this method can easily miss smaller features when they are placed on a larger feature. The method calculating the number of peaks does not have this weakness.

A common weakness of all but the ratio method, is the fact that step edges with complex features were somewhat transformed in order to be able to analyse them. Most features are not changed and from the features that are transformed, main characteristics like amplitude and frequency are mostly preserved

Chapter 5

Conclusion and recommendations

Substrates were cleaned with acetone and ethanol, and annealed at 1000°C for 4 hours. Measurements on as received and annealed DyScO₃ (110) substrates with XRD and AFM, have led to the conclusion that under the used circumstances, only a marginal connection between the in-plane miscut and frequency of features of step edges is present. A low in-plane miscut resulted in step edges with relatively low feature frequencies. Both in amplitude and complexity of features, no relation with the in-plane miscut is found. In relation to the polar miscut, a marginal connection between polar miscut and the amplitude of features can be distinguished. When the polar angle increases, the amplitude of features appears to decrease. Variation explained by in-plane and polar miscut is small, and additional factors are necessary to explain the variation of features of step edges in the extended dataset.

The methods of analysis in the present state provide a useful analysis, offering both a very basic detecting of complex features and a further analysis of amplitude and frequency of features. The ‘ratio’ method provides the basic detection of complex features, using the ratio of ‘experienced steps’ perpendicular to step edges to the number of step edges. Methods aimed at the amplitude measure the deviation from a straight line or 6th or 12th degree polynomial, and use a fourier transform in order to determine the deviation from a straight line. Methods aimed at measuring the frequency of features use the number of crossings of a straight line along a step edge or the number of peaks on a step edge as a measure for the average frequency. The methods combined give a useful representation of both amplitude and frequency of features on the step edges, and proved useful in analysing the results from the AFM measurements. However, both the method using 6th and 12th degree polynomials and the method using a fourier transform need to be expanded.

Recommendations

Several recommendations can be made in order to improve and guide future research. Future directions for research are advised, as well as possible improvements for both measurements and methods of analysis.

Because the influence of the miscut on the surface morphology is only marginal, there necessarily are other, more dominant, factors influencing the surface morphology of the DyScO₃ substrates. These factors possibly need to be sought for in differences between different batches of samples. In the batch used for the original dataset no samples with high levels of features were found, because two out of the three samples added in the extended dataset originate from the same batch. It would be interesting to obtain more information about the process of fabrication of the samples and to find out whether differences between samples/batches could originate from influences in the process of fabrication. The surface of samples with a high level of features, looks similar to substrates with a low polar miscut angle. Because the features on samples with low polar miscut are probably caused by atoms not being able to diffuse into the step edges due to the larger terrace width, it is natural to look for a diffusion related cause. Investigating the presence and influence of factors related to diffusion would be interesting from this point of view. It appears as though there is a factor holding back diffusion, other than temperature, anneal time and terrace width. The influence anneal temperature and time would however still be interesting to study, for example in order to get a better idea of the effects of long anneal times or high anneal temperatures on complex features. AFM images of samples

could be made at different points of the anneal process, in order to investigate the influence of anneal time on the presence of features.

While the analysis in this report was mainly aimed at the step edges, it could also be useful to study other parameters, like the surface termination and stoichiometry of substrates. Information about both can be collected using the phase information gathered with AFM, Time-of-Flight Mass Spectroscopy, surface x-ray diffraction or thin film growth.

The orientation of the surface in XRD was acquired using ζ and ξ scans at a low incident angle. This method however, uses a different part of the surface for determining ζ and ξ . In order to get a more reliable measure of the surface orientation, a laser may be used. While shining a laser at the centre of the sample, the sample is rotated. During this rotation, the location of the reflection from the laser of the sample surface is monitored. When the surface lies exactly in the same plane as the rotation, the reflection will not move during this rotation. This knowledge can be used in order to determine the orientation of the surface. Potential weaknesses of this method are the fact that, in order for this method to be reliable, the laser has to reflect from exactly the same spot during the entire rotation, because of the large variation in orientation on different locations of the surface. This measure is also most likely to give a local measure of the orientation (dependent on the diameter of the laser beam).

Most TM-AFM images used for this report were created with a scanning angle of 0° . This results in images that need to be rotated and cropped in order to be analysed. With the cropping, part of the image, and thus part of the information, is removed from the analysis. It would be advisable to use a scanning angle such that step edges lie vertical on the resulting TM-AFM image. This would minimize the need for rotating and cropping the images.

The methods of analysis developed mainly focus on the amplitude and average frequency of features. It would be advisable to expand the analysis in order to give a better measure of present frequencies within the features. The presence of certain frequencies could possibly provide information about the presence of certain types of features. Several possibilities exist to improve the measure of frequencies. First of all, the method using a 6^{th} and 12^{th} degree polynomial can be used. The higher the degree of the polynomial used to fit an edge, the better the fit will be able to approximate complex features. When two images give the same result for the 6^{th} degree polynomial, and a different result for the 12^{th} degree polynomial, this could implicate the image with the lowest deviation from the 12^{th} degree to have features fitted better with a 12^{th} degree polynomial than the other image. When this is done at a range of degrees, it could provide information regarding the complexity of the features, and thus regarding present frequencies. Another possibility using a polynomial would be to simply use a polynomial of a high order (± 50), and evaluate the resulting coefficients.

A more straightforward method of analysing present frequencies in the step edges, is a further analysis of the results from the method using a Fourier transform. The resulting power spectrum contains information regarding the presence of frequencies within the step edges. A further analysis of this result may yield a better understanding of frequencies present in step edges with a high level of complex features. Next to this expansion, it is also necessary to scale this method. As mentioned before in section 4.3, the result of this method is dependent on the length of the edges reviewed. In order to scale this method, the relation between the length of step edges and the result has to be determined.

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Appendix A

Comparison of AFM and XRD miscut measurements

Sample	Locatie	θ_v XRD ($^\circ$)	ϕ XRD ($^\circ$)	θ_v AFM ($^\circ$)	ϕ AFM ($^\circ$)
DSO03	center	0.15	34.66	0.16	31.31
	right top	0.15	34.66	0.19	41.63
	left top	0.15	34.66	0.19	35.66
	left bottom	0.15	34.66	0.16	28.29
	right bottom	0.15	34.66	0.14	24.28
DSO04	center	0.19	69.41	0.18	65.98
	right top	0.19	69.41	0.20	69.52
	left top	0.19	69.41	0.20	62.80
	left bottom	0.19	69.41	0.17	57.10
	right bottom	0.19	69.41	0.16	66.75
DSO05	center	0.16	68.44	0.17	67.93
	right top	0.16	68.44	0.22	78.64
	left top	0.16	68.44	0.20	56.94
	left bottom	0.16	68.44	0.18	39.56
	right bottom	0.16	68.44	0.09	63.72
DSO08	center	0.18	36.07	0.18	44.62
	right top	0.18	36.07	0.18	32.88
	left top	0.18	36.07	0.13	40.65
	left bottom	0.18	36.07	0.20	52.27
	right bottom	0.18	36.07	0.21	40.34
DSO11	center	0.16	-54.69	0.18	-57.93
	right top	0.16	-54.69	0.15	-58.90
	left top	0.16	-54.69	0.16	-48.27
	left bottom	0.16	-54.69	0.22	-63.57
	right bottom	0.16	-54.69	0.19	-67.07

Table A.1: Measurements of miscut compared between XRD and AFM, part 1, part 2 is Table A.2 on the next page.

Sample	Location	θ_v XRD ($^\circ$)	ϕ XRD ($^\circ$)	θ_v AFM ($^\circ$)	ϕ AFM ($^\circ$)
DSO12	center	0.2	-53.62	0.17	-57.03
	right top	0.2	-53.62	0.21	-54.91
	left top	0.2	-53.62	0.19	-62.76
	left bottom	0.2	-53.62	0.16	-53.07
	right bottom	0.2	-53.62	0.18	-44.70
DSO13	center	0.16	47.01	0.17	58.81
	right top	0.16	47.01	0.18	68.89
	left top	0.16	47.01	0.19	60.36
	left bottom	0.16	47.01	0.16	53.54
	right bottom	0.16	47.01	0.14	63.74
DSO14	center	0.09	-7.69	0.15	-6.65
	right top	0.09	-7.69	0.18	-12.30
	left top	0.09	-7.69	0.13	-20.75
	left bottom	0.09	-7.69	0.14	1.12
	right bottom	0.09	-7.69	0.17	-2.15
DSO17	center	0.13	0.59	0.14	-5.40
	right top	0.13	0.59	0.16	-12.06
	left top	0.13	0.59	0.14	-17.90
	left bottom	0.13	0.59	0.13	5.33
	right bottom	0.13	0.59	0.16	3.92
DSO19	center	0.12	-44.55	0.12	-52.14
	right top	0.12	-44.55	0.15	-45.06
	left top	0.12	-44.55	0.13	-61.41
	left bottom	0.12	-44.55	0.10	-51.14
	right bottom	0.12	-44.55	0.10	-40.34

Table A.2: Measurements of miscut compared between XRD and AFM, part 2.

Appendix B

Main parts of MATLAB scripts used for analysis

B.1 Giving the step edges a new x-variable (partial code)

```
for c=1:numel(Se) %follows every step edge one by one
    z{c}=1;
    x{c}(1)=2;
    y{c}(1)=Se(c);
    %c
    k=0;
    for w=1:1000 %follows one complete step edge
        %w
        if k==1 || x{c}(z{c})==m-1
            break
        else
            z{c}=z{c}+1; % the new x-coordinate
            d=0;
            f=1;
            clearvars places
            for r=1:100 %the distance from the present point to where it is looking for the next
                if d>0
                    x{c}(z{c})=(reserve{1}(1));
                    y{c}(z{c})=(reserve{1}(2));
                    break
                else
                    for th=1:360 %draws a circle to look at for every r
                        xr(th)=round(0.2*r*cos(th*pi/180));
                        yr(th)=round(0.2*r*sin(th*pi/180));
                        xn(th)=x{c}(z{c}-1)+xr(th);
                        yn(th)=y{c}(z{c}-1)+yr(th);
                        if xn(th)<1
                            xn(th)=1;
                        end
                        if yn(th)<1
                            yn(th)=1;
                        end
                        if yn(th)>n
                            yn(th)=n;
                        end
                    end
                end
            end
        end
    end
end
```

B.2 Calculating the ratio of ‘experienced steps’ perpendicular to step edges to the number of step edges.

```
%ratio after edge, crosses the sample with a 90 degree angle compared to
%the stepp edges, looks for the amount of steps it crosses, and compares
%this to the amount of steps in the sample
function [Ratio]=fratioedge(filename,Np,thresh,sigma)
data=importdata(filename);
[m,n]=size(data);
enen=edge(data,'canny',thresh,sigma)+0;
%imshow(enen)
c=1;
for x=2:m-1
    [pks,lcs]=findpeaks(enen(x,:));
    Nc(c)=numel(lcs);
    c=c+1;
end
ANc=mean(Nc);
Ratio=ANc/Np;
end
```

B.3 Calculating the standard deviation from a straight line (partial code)

```

for e=1:numel(Se)
    clearvars f
    x{e}=1:numel(y{e});
    p{e}=polyfit(x{e},y{e},1);
    c=p{e}(1);
    d=p{e}(2);
    f(1)=c+d;
    for b=2:numel(y{e})
        f(b)=c*b+d;
        if (f(b)>y{e}(b) && f(b-1)<y{e}(b-1)) || (f(b)<y{e}(b) && f(b-1)>y{e}(b-1))
            end
        end
    end
    z{e}=y{e}-f;
    s(e)=std(z{e},1);
end
msd=mean(s)*scale; %micrometer
end

```

B.4 Calculating the number of crossings of a straight line along the step edges (partial code)

```

for e=1:numel(Se)
    nc(e)=0;
    x{e}=1:numel(y{e});
    p{e}=polyfit(x{e},y{e},1);
    c=p{e}(1);
    d=p{e}(2);
    f(1)=c+d;
    for b=2:numel(y{e})
        f(b)=c*b+d;
        if (f(b)>y{e}(b) && f(b-1)<y{e}(b-1)) || (f(b)<y{e}(b) && f(b-1)>y{e}(b-1))
            nc(e)=nc(e)+1;
        end
    end
    z{e}=y{e}-f;
end

mnc=mean(nc)/(m*scale); %1/micrometer

```

B.5 Calculating the standard deviation from a 6th and 12th degree polynomial

```

function [sdpol,sdpol2X]=fpolynom(y,degree) %fits the steps of a sample with a 'degree' 2X'degree'
%polynomial and subtracts this polynomial from the steps. Finally it calculates the standard
% deviation of the result, for both polynomials.
close all
clearvars df sd sdpol
s=warning('off','MATLAB:polyfit:RepeatedPointsOrRescale');

```

```

for a=1:numel(y)
    clearvars df
    h{a}=[1:numel(y{a})];
    p{a}=polyfit(h{a},y{a},degree);
    f=polyval(p{a},h{a});
    df=y{a}-f;
    sd(a)=std(df,1);
end
sdpol=mean(sd);
for a=1:numel(y)
    clearvars df sd
    h{a}=[1:numel(y{a})];
    p2X{a}=polyfit(h{a},y{a},2*degree);
    f=polyval(p2X{a},h{a});
    df=y{a}-f;
    sd(a)=std(df,1);
end
sdpol2X=mean(sd);
warning(s);
end

```

B.6 Calculating the number of peaks of step edges

```

function [nps]=fpieken(y,m,scale)
for a=1:numel(y)
    pks{a}=findpeaks(y{a});
    nps(a)=numel(pks{a});
end
nps=mean(nps)/(m*scale);
end

```

B.7 Calculating the integral of the power spectrum

```

function [ints,ath]=ffrequency(y,scale) %calculates the power spectrum for all steps apart and
%sums this in order to have one grand total for a sample
for c=1:numel(y)
    y{c}=y{c}-mean(y{c});
    x=1:numel(y{c});
    p=polyfit(x,y{c},1);
    pf=polyval(p,x);
    y{c}=y{c}-pf;
    Fs=abs(fft(y{c}))/(numel(y{c})/2);
    Fs=Fs(1:(numel(y{c})/2)).^2;
    lf=[0:(numel(y{c})-2)/2]/256;
    theta(c)=atan(p(1));
    if theta(c)>pi
        theta(c)=theta(c)-2*pi;
    end
    totint(c)=trapz(lf,Fs);
end
ints=sum(totint);
end

```