PIEZORESPONSE FORCE MICROSCOPY USING ADAPTIVE CONTACT-MODE IMAGING

By

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ABSTRACT OF THE THESIS

Piezoresponse Force Microscopy using Adaptive Contact-Mode Imaging

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Atomic Force Microscopy (AFM) is a type of Scanning Probe Microscopy (SPM) which involves a probe tip scanning over a sample surface for obtaining data of the sample. Data acquisition is carried out by making physical contact with the surface of the sample, thus there is scope of characterizing data even beyond basic topology. Piezoresponse Force Microscopy (PFM) is just an extension of AFM imaging which is used for obtaining the piezoelectric response of the sample using the converse piezoelectric effect. Piezoelectric effect can be observed in almost all the materials around us which include biological and inorganic materials. PFM imaging is extensively used in the nanoscale characterization of ferroelectrics and its applications continue to grow. Thus, with the growing use of this technique, it is important to obtain images of high quality and at a faster rate to reduce the time required to image without affecting its accuracy. Research and experiments show that with the help of different algorithms and external control, the speed of AFM imaging can be boosted with reduced loss of data and higher accuracy. This research aims at applying existing control methodologies used in AFM imaging to the PFM imaging and analyzing the output by comparing images obtained at lower and higher speeds using different methods.

Dedication

I would like to dedicate this thesis to everyone who has supported me through every step that I took. Firstly, I would like to dedicate this to my mother Megha and father Mahesh for their constant and unconditional love and support. I would also like to dedicate this to my family which includes my brother, uncles, aunts and cousins for always being there whenever I needed them.

This dedication is also for my friends here and back home for all the help and support they provided and for being the only family that I had far away from my home.

I would also like to dedicate this to all my colleagues and professors who may have directly or indirectly helped me through my journey. Lastly, I would like to dedicate this to Dr. Zou and all the members of the laboratory for making it easier to understand my work and helping me whenever required as this would not have been possible without them.

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Chapter 1

Introduction

Over the past few decades, the development in the nanoscale imaging has been instrumental in understanding and studying the fundamental characteristics of materials. Development of the technique of Scanning Probe Microscopy serves an important role in fast and accurate data acquisition. An Atomic Force Microscope (AFM) is one such device which has high resolution characterization of virtually all type of materials. The AFM is a type of scanning probe microscope which obtains topological data of the sample by scanning a probe over it. As the operation of AFM is mechanical in nature, it just requires a physical contact with the surface of the sample, which is why it can be used on even biological samples. The main principle of detection of topography is that the scanning probe tip has a laser trained over it, which displaces with the displacement of the probe in either lateral or vertical direction. This change in laser position is converted into deflection signal which can be read out through a software application along with the position of the height sensor. Thus, a 3-dimensional image of the sample can be obtained in a non-destructive manner. Apart from detecting just topology, the AFM has numerous modes fine-tuned for obtaining different kinds of information, which include other mechanical and electrical characterization. One such mode is the Piezoresponse Force Microscopy mode. When operated in this mode, the AFM is called as a Piezoresponse Force Microscope (PFM) and is utilized for nanoscale mapping and manipulation of domain structures in ferroelectric materials.

PFM modifies the normal operation of AFM by applying a bias at the tip-sample connection. This method is based on the detection of local piezoelectric deformation of a suitable sample using an external electric field. It can only be used on materials which exhibit the property of piezoelectricity. The external AC bias induces a deflection of the sample which is of a very low magnitude (few nanometers) which is detected by the AFM and later processed through a Lock-in amplifier (LIA). The LIA is built in inside the AFM which separates the PFM signal from the topology data and the background noise using the frequency of the input AC bias. The nanometer resolution of AFM is very useful to accurately map the deflection due to piezoelectric effect which is usually in that range. The study of PFM is essential for fast and accurate characterization of piezoresponse and finds it use in a number of applications. PFM is extensively used in the studies of domain poling, switching mechanism in ferroelectric structures and hysteresis behavior Gruverman and Kalinin [2006]. The rapid growth in the use of devices using ferroelectric materials has created a strong need to investigate the properties of these materials at the nanoscale resolution Gruverman and Kalinin [2006]. Thus, the development of better quantification of data using PFM in terms of the spatial resolution and domain dynamics has been given a lot of importance (Soergel [2011]). There is always a need to study ways to acquire data at a faster rate without reduction in the quality of the data and without modifying the sample's physical and electrical properties (Nath et al. [2008]).

This paper presents an approach which has been successfully applied at obtaining better topological images using a standard AFM. This utilizes an external control with an algorithm which applies Adaptive Contact-Mode (ACM) approach Ren and Zou [2014]. This approach uses feed-forward and feedback mechanisms to better locate the probe tip on the surface of the sample without compromising on the scan rate and with better accuracy. The ACM technique minimizes the contact force between the tip and the sample and maintains sufficient contact between the two. When applied to PFM, it can be used to operate at higher scan rates with better topology and piezoresponse data capture.

The structure of this paper involves understanding the basic principle of working of the AFM and the PFM by understanding the concept of Piezoelectricity and it also explains the need of a PFM to accurately characterize ferroelectrics at nanoscale resolution. We then move to the procedure for performing experiments on a standard sample and explaining the interpretation of results. Later, we present a mechanism for operating the PFM at a higher scan rate and obtaining better data acquisition using the Adaptive Contact-Mode Imaging approach. At the end, we compare the images acquired by standard software and external control to show how the external control gives better data acquisition at a faster rate.

Chapter 2

Literature Review

– read and restructure –

The Atomic Force Microscope is a mechanical microscope which can obtain surface information of the samples. It cannot replace an optical microscope, but being mechanical in nature, can be used on various materials ranging from hard metals to soft cells. Initially, AFM was used for characterizing surface of non-biological materials, but today finds most of its application in biological and biomedical research. Chang et al. [2012]

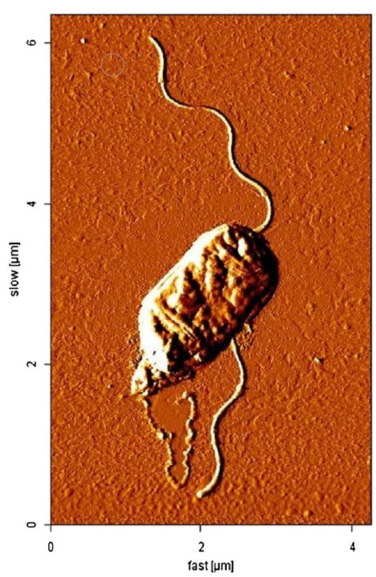

Figure 2.1: Image of an E-coli using an AFM Chang et al. [2012]

The preparation of samples for AFM experiments does not require special procedures and the AFM is also capable for operating in air and liquid. Thus, samples can be scanned in their physiological buffer solutions Chang et al. [2012]. The AFM finds its application in the field of virology as it is able to scan virus like Moloney murine leukemia Kuznetsov et al. [2004], HIV Kuznetsov et al. [2003] or plant cells in their crystalline form Malkin et al. [2002]. A lot of work can also be found in DNA and chromosome studies Rippe et al. [1997], visualization of the surface of bacteria Miller et al. [2006], imaging of live mammalian cells Braet et al. [1998] and in cell morphology Kim et al. [2012].

The scanning of images is performed at a certain speed, the factors for which are determined by the physical limitations of the sample and the probe. These factors mainly arise due to the physical nature of the contact between the tip and the sample. A major part of research involves operating the AFM at faster scan rates with the same nanoscale resolution output. The maximum scan speeds may be determined by the physical properties of the probe, like the stiffness of the cantilever (Butt et al. [1993]), or the maximum amount of force that can be applied on the sample. As AFM is used with vast type of sample, researchers have come up with various solutions for attaining high speeds for scanning, like integrating actuator and sensor in the cantilever (Manalis et al. [1996]) or by using a microresonator as a scan stage (Humphris et al. [2005]). Improvisations have also been made using algorithms for maximizing tip-sample contact and maintaining limited contact force using external control algorithms.

Some external control techniques which have shown efficient tracking are Iterative Control techniques. A Model-less Inversion based Iterative Control technique which does not require a dynamic model of the system, was shown to produce precision output tracking even with some disturbance or noise present in the system (Kim and Zou [2008]). Another approach, called adaptive contact mode (ACM) technique, uses quantification of topographical data to provide a feedforward control and achieve precision tracking of the sample topography (Ren and Zou [2014]). These two methods are used for high-speed scanning and are further discussed and utilized for external control in this paper to increase the scan speed and obtain precision tracking of the sample. The ACM technique will be applied to the Piezoresponse Force Microscopy for high-speed scanning and results will be compared with the standard high-speed scanning.

Piezoelectricity is a form of coupling between electrical and mechanical property of a material. It can be found in inorganic, organic or even biological systems. This property enables us to obtain electrical polarization upon application of mechanical stress, or conversely change the structure of the material by application of external electrical signal. PFM enables use of this converse effect to map the amount of piezoelectric deformation and polarization direction of a material which is electromechanically active. This coupling was found to exist in almost all the systems around us as reviewed by Kalinin et al. [2006] who wrote that a variety of biological systems like the bones, teeth, wood or even cells, proteins and muscles are found to have electromechanical property.

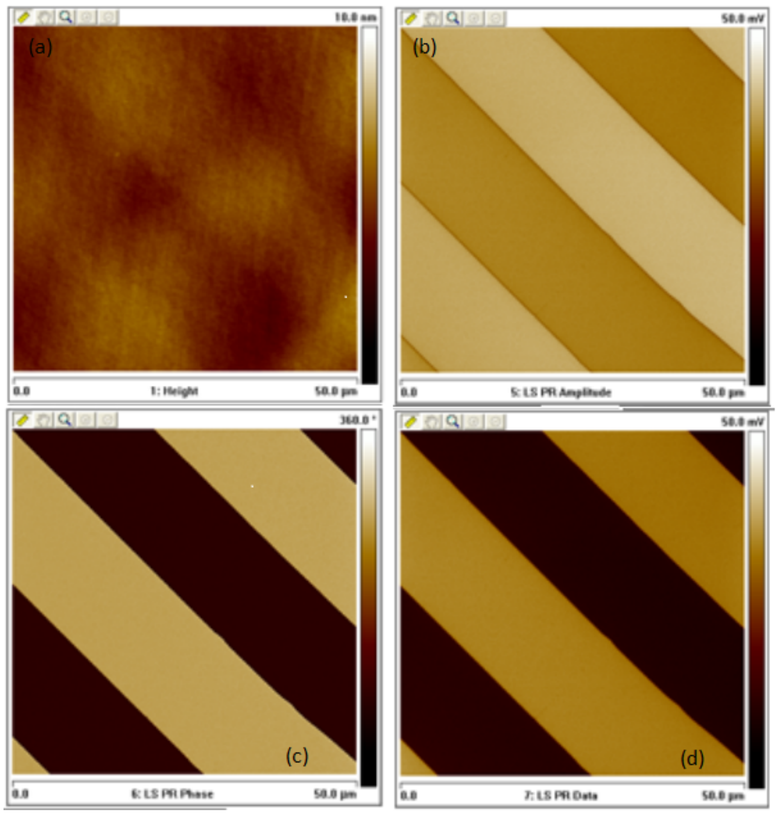

Figure 2.2: PFM image of a Periodically Poled Lithium Niobate sample. (a) Height sensor data, (b) PFM Amplitude data, (c) PFM Phase data and (d) PFM Sum

The study of Piezoresponse Force Microscopy is carried out over a large domain which includes piezoelectric and ferroelectric materials. These systems contain an active electro-mechanical structure which can be modified. Over the years, study has been performed to improvise the mechanism of imaging, to find faster and more efficient way to get an output without affecting the sample under test. One of the major areas of study has been in the mapping of ferroelectric surfaces (Bonnell et al. [2009])

The High Speed Piezoresponse Force Microscopy (HSPFM) carried out by Nath et al. [2008] shows an area of study which involves imaging of samples at a faster rate for allowing dynamic study of domain structure. Taking advantage of the smoothness of the sample, let them eliminate the use of feedback mechanism which mainly aimed at reducing tip-sample interaction caused in a rough sample. This technique is particularly useful with samples having a smooth surface, as for rough sample, the tip-sample interaction plays an important role in limiting the scanning speed. The technique presented in this paper aims at reaching higher scan rates for smooth and rough samples without compromising on the output image quality. Reaching each and every pixel and obtaining data from it is the main idea behind this method, which was previously employed on a standard AFM to obtain accurate topological information.

Chapter 3

Piezoresponse Force Microscopy

Piezoresponse Force (PF) Microscopy is a Scanning Probe Microscopy technique which is an extension of Atomic Force Microscopy. It has been developed since a couple of decades and predominantly finds its use in characterizing ferroelectrics. Since this technique was first demonstrated Güthner and Dransfeld [1992], it has become a standard for imaging ferroelectric domain patterns. It is based on the detection of local piezoelectric deformation of a ferroelectric sample induced by an external electric field. This chapter provides details about PF microscopy, with its applications, experimentation and results interpretation.

1. **Ferroelectrics:**

   For understanding the working principle of a PFM, we need to understand the concept of piezoelectricity and ferroelectric domains in the context of a PFM. A ferroelectric material is that which has a spontaneous electrical polarization that can be reversed upon the application of an external electric field Lines and Glass [2001]. All ferroelectric materials are pyroelectric and piezoelectric. Thus, all ferroelectric materials exhibit the property of piezoelectricity which is why we are able to characterize such materials using a PFM.

2. **Piezoelectricity:**

   The piezoelectric effect is the electric potential generated across a material when a mechanical stress is applied to it. This effect arises due to the asymmetry in the unit cell of a crystal which generates a polar effect. The application of mechanical stress causes shifting inside such unit cells which align themselves in a direction causing a flow of electric current. Inversely, when an electric potential is applied across a piezoelectric material, the direction of the poles changes which in-turn change the shape of the structure.

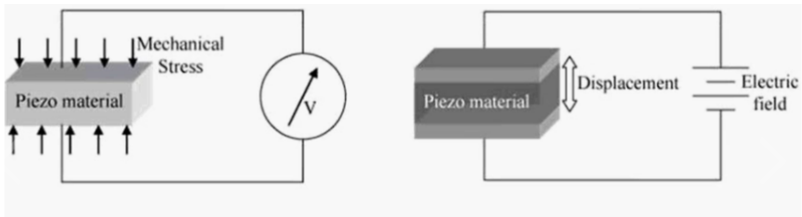

Figure 3.1: Piezoelectric and Inverse Piezoelectric effect

The deformation of the structure can be an elongation, contraction or shear depending on the direction of the applied field and the direction of polarization of the sample itself. In tensor form, the effect can be explained in terms of the polarization (P), the piezoelectric co-efficient (d_{ijk}) and the stress applied (σ)

$$P = d_{ijk} * \sigma \tag{3.1}$$

Conversely, application of uniform electric field along the polarization direction results in the elongation or contraction of the domain depending on its polarization direction with respect to the direction of the applied field. The field induced strain is given by

$$\begin{aligned} S &= \frac{\Delta Z}{Z} \\ &= \pm d_{33} * \epsilon \end{aligned} \tag{3.2}$$

This can be rewritten as

$$\Delta Z = \pm d_{33} * V \tag{3.3}$$

Where, V is the applied voltage, the $\pm$ sign reflects the direction of the domains, which helps in analyzing opposite domains.

3. **Ferroelectric domains:**

A ferroelectric domain can be defined as an area in which the spontaneous polarization direction points in a single direction. A domain wall separates two adjacent domains. A PFM can successfully map separate domains in a ferroelectric with nanoscale precision which makes it a very useful tool in characterizing such structures.

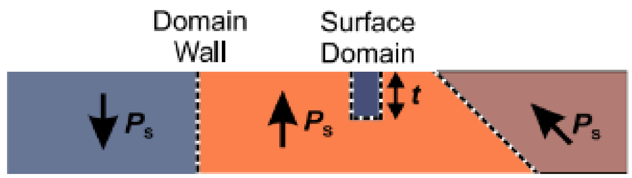

Figure 3.2: Ferroelectric domains [Soergel [2011]]

4. **Experimental Basics:**

As stated earlier, a PFM is a type of Scanning Probe Microscope which is just an extension to the Atomic Force Microscope. To truly understand the working of a PFM, we need to know the basics of an AFM and how the effect of piezoresponse is obtained through some additional changes to the normal AFM operation.

(i) **Atomic Force Microscope:**

An AFM is a scanning probe microscope in which a probe with a fine tip scans over the surface of the sample to obtain its topological data. The probe material is selected according to the sample surface to be scanned. The movement of the tip is controlled in 3 linear axes with a piezo-sensor on the z-axis which is used for feedback control of the tip for changes in the sample surface in the z-direction. The sample is mounted on a platform which can only be moved in lateral X and Y direction. A software application enables positioning of the sample, focusing the probe on the area which needs to be scanned, focusing the laser on the tip and viewing the sample scanning in real time.

The probe is situated on a cantilever which rests on a probe holder. A laser beam is trained on the back of the cantilever which reflects back on to a mirror which sends it to a photo diode. Any deflection in the movement of the probe causes the laser beam to change its position thus generating an error in the feedback loop. Initially, when the probe is just in contact with the sample, due to the interatomic forces there is a pull towards the sample, Thus when there is a dip in height in the sample, the probe loses contact and there is movement in the laser beam. This error is minimized using a feedback control on the Z-piezo which moves the probe downwards to reinitiate contact with the sample. The same methodology works when there is an increase in height in the sample, thus, the feedback control ensures constant contact between the probe and the sample. Due to the constant contact, the topology mapping of the sample surface can be done accurately down to the size of a few nanometers.

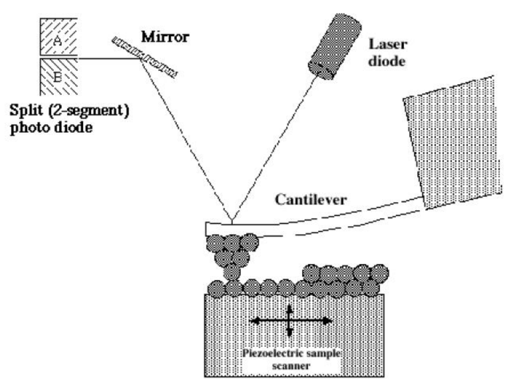

Figure 3.3: Optical lever detection of Cantilever deflection

The common modes of operation of an AFM are contact mode and tapping mode. These are classified based on the type of contact between the probe and the sample. In contact mode, the tip of the probe is in constant contact with the sample surface. The tip moves in the vertical direction according to the surface topography. This mode tracks the surface topography very well for larger variation in height. However, this mode results in larger lateral forces, which can be avoided by operating it in tapping mode. Tapping mode is an intermittent contact mode in which the tip oscillates at its resonant frequency and is in contact with the surface only at the lower part of its oscillation (Gadegaard, 2009).

The sample needs to be prepared before it can be used on the microscope. This depends on the type of sample and the modes of operation. Because AFM is a mechanical microscope, even biological samples can be mapped which need to be kept inside a fluid during when there is contact between the tip and the sample. The only limitation for surface mapping is its size. The AFM has limited range of movement is X, Y and Z direction with the Z-range being typically between 1-20 μm.

The images obtained can be analyzed using software application. When using external control, the data can be extracted using Signal Access Module and a plot can be obtained for the deflection or the Z-sensor movement for change in lateral movement of the probe, thus generating a 3-Dimensional profile of the topology of the sample under consideration.

(ii) **Piezoresponse Force Microscopy:**

Piezoresponse Force Microscopy is a type of Scanning Probe Microscopy which uses a standard AFM operated in contact mode with an additional voltage applied to the conductive tip. This causes periodic deformation of the sample surface which is transmitted to the tip. A lock-in amplifier then reads out the oscillations by cancelling out the noise in the output.

The piezo-electric effect causes the material to deform depending upon the direction of polarization and the applied electric field. Typically, the piezo-electric coefficient of the sample lies in the pm/V range which is very small to detect, especially when we have a background noise. Thus, applying a voltage induces a deflection which cannot be detected accurately. Applying an AC bias gives the input and the output a frequency signature which help in singling out the PFM signal from the background noise using a lock-in amplifier. Depending on the frequency of the AC bias, we can operate the lock-in amplifier at low and high speed.

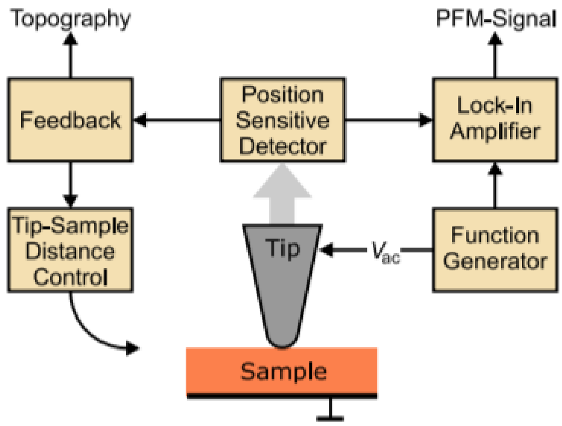

Figure 3.4: Schematic of PFM as an extension to AFM

Due to its extremely high vertical sensitivity, the AFM can measure topography variations in the nano-scale regions. PFM simply modifies the basic AFM operation by applying an AC Bias across the tip and the sample at a frequency. The output obtained is in the form of PFM amplitude and phase with the help of a lock-in amplifier. The LIA compares the output with an external reference frequency to extract the useful output from a noisy background.

The main factors affecting the output of the PFM are the frequency and the external alternating voltage. For a standard PFM, the frequency f of the AC Voltage V is set to values ranging from 1-100 kHz and the amplitude ranging from 1-10 V. The frequency should be high enough to not affect the simultaneous recording of the topological data of the sample, while it should also be small enough to avoid contact resonance of the cantilever. According to the equation 3.3, the PFM signal is a function of Voltage applied. Higher the voltage, larger is the value of the PFM signal. But, due to small tip radius, there is a chance of extremely high local electric fields which may modify the domain pattern by local poling. Thus, there is a restriction on increasing the applied voltage on the sample.

5. **Interpretation of Results:**

The operation of PFM is fairly similar to that of an AFM with an additional external AC Voltage applied on the sample or the tip. Hence, the typical results obtained are the topography of the sample, the deflection error, the amplitude of the PFM signal and the phase. The first two images are obtained from standard AFM operation while the next two are related to the PFM. The PFM amplitude is simply the deflection due to the application of the external voltage. It depends on the piezoelectric coefficient of the surface of the sample under test. For applications where the sample contains periodic poling or domains with different concentrations of dipole, we obtain a contrast in the images for the amplitude and the phase. The phase provides data regarding the alignment of the poles upon application of external AC bias which can be either 0° for in-phase or 180° for out of phase alignment.

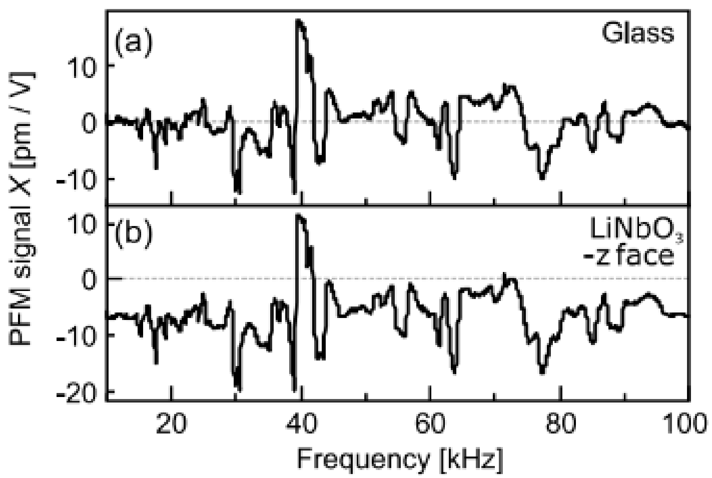

Figure 3.5: Similar PR signals for a non-piezoelectric and piezoelectric sample

The contrast in the images for the amplitude and the phase largely depends on the selection of the frequency f (Labardi et al. [2000]) and the AC voltage V applied 3.3. Another major factor affecting the PFM contrast is the system background noise. This signal cannot be eliminated and is due to the complex mechanisms of the AFM head. The effect of background can be shown by using the PF Microscopy on a non-piezoelectric sample and then on a standard one. The figure shows that the signature of the PFM signal is exactly the same even if the glass is not a piezoelectric material, due to the fact that the background is generated by the system itself and not by the sample. A background free imaging is thus required for accurate data acquisition using the PFM.

6. **Applications of PF Microscopy:**

A PFM can probe electro-mechanical response from materials ranging from ferroelectric and piezoelectric samples to biological tissues. The nanoscale precision is useful in accurately quantifying the data, and the constant work in improving the quality of data acquisition has led to many useful applications for the PFM. The study of ferroelectric domains is largely carried out with the help of a PFM. An accurate mapping of piezoresponse obtained Ferroelectric materials can be found in a large number of products in the real world. Thus, the study of domains and its different aspects with nanoscale precision with the help of PFM has been given a lot of importance. Some of the more detailed applications are illustrated below (Bonnell et al. [2009]):

- Ferroelectric materials contain a number of possible defects in their structure that influence the local ferroelectric switching. Also, the ferroelectric domain are separated by domain walls which may interact with the local defects resulting in further defects. The study of such materials enables us to identify defects on the surface of the sample along with the possible location of the domain walls. All this at a nanoscale precision goes a long way in further understanding the structure of the sample without affecting its use.
- The polarization dynamics in ferroelectric thin films and capacitors can be observed using PF Microscopy. A global excitation approach can be applied to emulate the switching condition in actual devices with integrated ferroelectric capacitors to allow quantitative studies of the nucleation and domain wall dynamics.

- PFM can also be employed to image the local property variations in piezoelectric semiconductors. The design and performance of GaN-based electric and optoelectronic devices can be improvised by studying their electromechanical response at nanoscale precision.

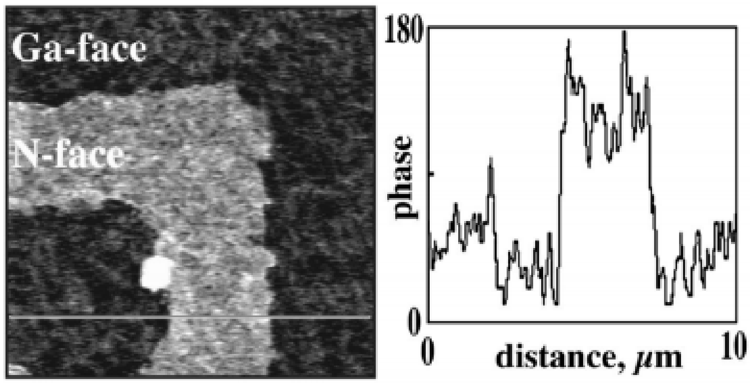

Figure 3.6: Image of PFM phase and its cross section of a GaN sample

- A variety of biological systems exhibit piezoelectric behavior including certain tissues and plants. The study of relationship between mechanical properties and the electric fields is the main idea behind using PFM for imaging such materials. The use of vertical and lateral PFM imaging further helps in studying complex mechanisms inside such materials

- The ability of the PFM to manipulate ferroelectric polarization has also had a large impact on the nanostructure fabrication and high-density information storage.

Chapter 4

High Speed Adaptive Contact Mode Piezoresponse Imaging

The PFM is typically run at speeds with which the AFM is utilized. Thus, time required to acquire an image is very high, limiting its use for dynamic studies of materials. This problem was also addressed by (Nath et al. [2008]) where they increased the speed of normal PFM operation but with the use of a smooth sample surface. The technique included no use of feedback to maintain tip-sample contact as the surface was smooth and without much topography variation. Working with rough surfaces without tip-sample interaction feedback is very dangerous as there is a chance of tip or the sample getting permanently damaged due to the high contact force. We also know (Jesse et al. [2006]) the importance of a good tip-sample contact (no potential drop across tip-sample gap) is important for accurate data acquisition. Thus, for high speed imaging on surface with topology variations, we need to maintain a constant tip-sample surface contact to ensure accurate data acquisition along and reduced tip-sample wear.

The tip-sample contact is maintained by the use of a feedback control which locates the probe tip relative to the sample and adjusts its location to maintain a constant contact. For a typical AFM operation, the PI gains are tuned to obtain stable contact and thus obtain precise images. PI control can be best utilized when used at lower speeds (scan rate $\leq$ 10 Hz). At higher speeds, however, the images for the topology that are obtained are not as accurate as there are good chances that the tip loses contact of the surface where topology changes suddenly. In case of PFM, losing the tip-sample contact would create a potential drop across the gap which would create problems in acquiring accurate and precise data. Thus, we need to increase the operating speed of the imaging without losing tip-sample contact and with minimal contact force. This can be done with the help of Adaptive Contact-Mode imaging technique (cite). This is a type of external control technique which applies its own set of feedforward and feedback mechanisms to accurately track the sample topology with minimum interaction force.

The increase in the scan speed with minimal interaction forces can be obtained from the following steps: (i) The sample topography was quantified using the height sensor as well as the deflection signal instead of only the height sensor. (ii) The deflection set point was adjusted line-by-line around a minimal level to maintain a stable tip-sample contact instead of a fixed deflection set-point and (iii) the sample topography quantified in the first part is utilized in an iterative feedforward control scheme to track the sample topography. For using this technique, it is required that the cantilever is not excited at its resonant frequency which is also a desirable state for the PFM imaging. During standard contact mode imaging at high speeds, the large deflections cause low quality height images due to the slow response of conventional control at higher speeds. This leads to erroneous results for the topography of the sample. Using this technique, the change or the difference in the height of any two points on the sample with height variance can be obtained by adding the deflection difference to the height difference.

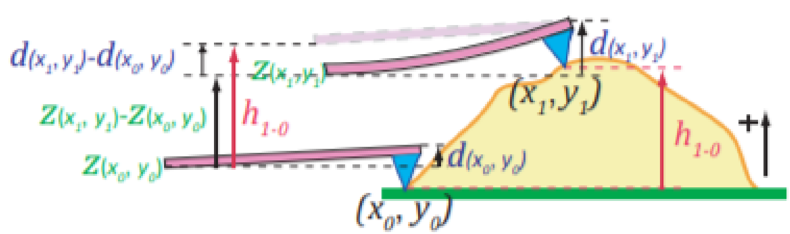

Figure 4.1: Difference in height at two locations on the sample and the deflection of the cantilever at each point

Consider the figure 4.1 where the datum is set at (x_0, y_0). The first point is at (x_0, y_0) where the deflection of the cantilever is $d(x_0, y_0)$ and the z-sensor is at $z(x_0, y_0)$. When we reach the point (x_1, y_1), the deflection of the cantilever is $d(x_1, y_1)$ and corresponding z-sensor position is $z(x_1, y_1)$. Thus, the actual difference in height between the two points is given by

$$h_{0-1} = [z(x_1, y_1) - z(x_0, y_0)] + [d(x_1, y_1) - d(x_0, y_0)] \tag{4.1}$$

We can set the datum to be $z(x_0, y_0) = 0$ and $d(x_0, y_0) = d_{set}$. Thus, for a general point (x, y) with respect to the datum, we obtain the following

$$h(x, y) = z(x, y) + [d(x, y) - d_{set}] \tag{4.2}$$

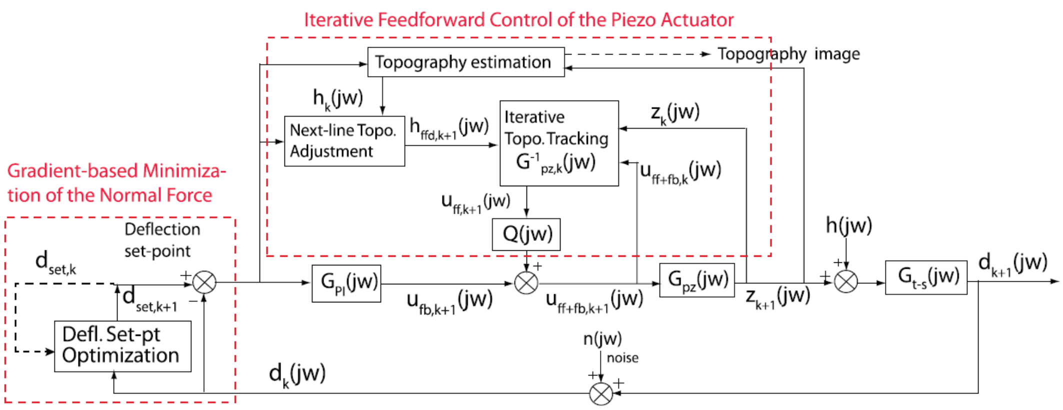

Figure 4.2: Block diagram for the Adaptive Contact Mode imaging

The equation (ref) suggests that the deflection set point need not be fixed during the imaging process. This enables us to reduce the contact force between the tip and the sample. The deflection set point can be adjusted line by line by using the quantified topology data obtained from the previous step. A gradient based optimization method was then applied to reduce the normal force on the sample and also maintain a stable contact. Subsequently, an iterative feedforward mechanism was used to control the z direction piezo. This was done by estimating the sample topography and the tracking error at the next scan line and using this estimation to control the z-direction piezo actuation for sample topography tracking.

This method has been successfully applied to the AFM and the speed of imaging was increased up to 30 times along with a substantial decrease in the interaction force. The figure 4.3 shows how high scan speeds provide images with almost the same precision if not better when using ACM imaging. PF Microscopy is just an extension of AF

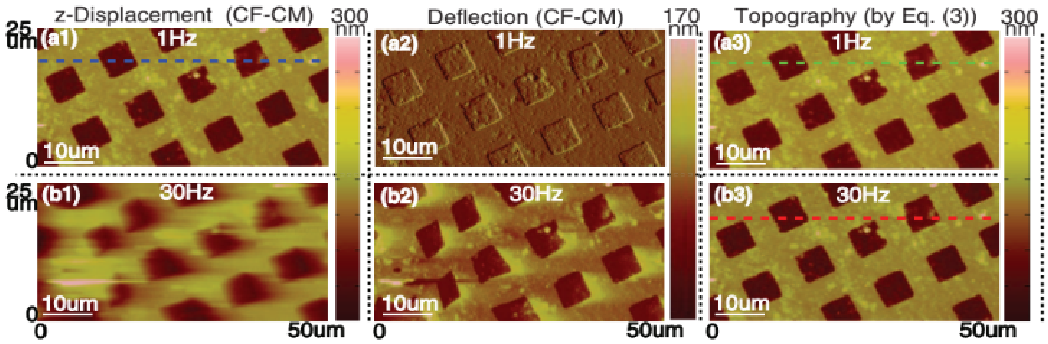

Figure 4.3: Images using conventional Contact Mode imaging (a1, a2 and a3) and Adaptive Contact Mode Imaging (b1, b2 and b3) (Ren and Zou [2014]

Microscopy, in which there is a similar requirement of maintaining contact with the sample without exerting too much force and maintaining nanoscale precision. Thus, integrating the high-speed ACM method with the PFM is a logical step towards effective high-speed PFM imaging.

Chapter 5

Experimental Procedure

The procedure for imaging using PFM is similar to that of a standard AFM. For the purpose of this experiment, we are using Dimension ICON with no other extra components and connections. This project aims at using an algorithm to perform the imaging at higher speeds with better data acquisition. Thus, one part of this project involves performing experiment using the standard Nanoscope software provided by Bruker to use the AFM in Piezoresponse mode. The next part involves use of an external control to operate the AFM at higher scan rate and obtain images. The images will then be compared to show which control suits better for imaging and the areas where external control can be improvised.

The set-up of the experiment to perform PFM imaging is by setting up the standard AFM with a Dimension ICON head. After setting up the ICON head, we move on the select a suitable probe for the experiment. As this experiment involves application of an AC bias, we need to select a probe with a conductive layer. Also, for better sensitivity, the cantilever length should be shorter and its spring constant should be higher. There are a few probes that fit into this criteria shown in the table 5.1

Probe	**Shape**	**Resonant Freq.** kHz			**Spring Const.** N/m			**Length** µm			**Width** µm		
		Nom.	Min.	Max.	Nom.	Min.	Max.	Nom.	Min.	Max.	Nom.	Min.	Max.
SCM-PIT V2	Rectangular	75	50	100	3.0	1.5	6.0	225	215	235	35	33	37
MESP-RC	Rectangular	150	100	200	5	2.5	10	125	115	135	30	25	35
DDESP V2	Rectangular	450	300	600	80	30	180	125	115	135	40	38	42
MESP V2	Rectangular	75	50	100	3.0	1.5	6.0	225	215	235	35	33	37

Figure 5.1: Table showing the different compatible probes for performing Piezoresponse Force Microscopy

For this experiment, we selected the SCM-PIT-V2 probe as it was already available.

After selecting the probe, we mount it on the scanner and tune the laser on the photodiode. We then install the sample at its position on the base. We used a Nickel doped (5%) Zinc Oxide sample manufactured using sputtering process. Once the sample was installed, we set initial parameters for scanning as shown in figure 5.2

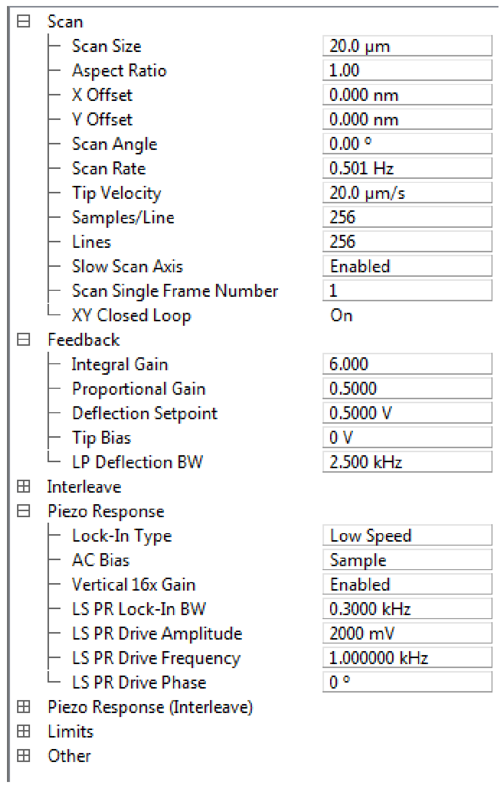

Figure 5.2: Scan parameters for PFM operation

Two main parameters for precise PFM imaging are the operating voltage and frequency. As stated earlier, the frequency at which the voltage is applied plays an important role in data acquisition. We need to set this frequency according to the probe and sample material to avoid conditions like contact resonance which may drastically change the output signal. The initial values chosen were $f = 1kHz$ and $V = 1000mV$. We can change these values once we start with the imaging for further tuning.

The most important part of imaging is that the tip must remain engaged with the sample at all times. Even if the sample is smooth enough, the sudden fluctuations needed to be taken into consideration while performing the experiment and the values for the feedback gains need to be adjusted accordingly. If the tip maintains a good contact with the sample surface, the piezoresponse can be obtained accurately as it has an active electrical connection.

The first part of the project is to run the PFM experiment and obtain images using the standard Nanoscope software. Thus, we simply set the parameters and engage the tip to start imaging. The images obtained are those of the surface topology, the deflection, the PFM Amplitude and the PFM Phase. These data channels can be set according to preference in the software. We can obtain real time images which helps in faster tuning of the parameters. After we engage, we first tune the PI gains to accurately trace the surface topology of the sample. We then move on to optimizing the PFM output. From the equation 3.3, we know that the Voltage across the sample is directly proportional to the deflection output. Thus, higher the voltage, larger the PFM signal but then there is a chance that high voltage will permanently polarize the sample which will provide inaccurate results along with damaged sample. The frequency which is selected, is also affected by a certain factors. When we come across a surface which does not react to the external voltage, or if the surface is not a piezoelectric surface, we do not expect any signal for the amplitude. Even though we do not have any surface deflection, we get a PR signal due to the background noise. This can be avoided by selecting suitable value for frequency at which we apply the voltage. As this, is not of the main concern for this project, we did not apply any optimization for the frequency selection.

The PFM data is acquired with the help of a lock-in amplifier which separates the output signal providing the values for PR Amplitude and corresponding Phase. Thus, with the help of the frequency provided by us and the lock-in amplifier, the PFM data is obtained. Same tests were run at different speeds for observing changes in the output using only the Nanoscope software. These results were later used for comparison with the results obtained by using external control.

The next step was to use external control for scanning. We need to make changes to the registers through the software to enable controlling externally. For this experiment, we used Matlab and XPCTarget, along with a DAQ card system to send and receive data to and from the AFM. Through the NS V Controllers front panel, we can send signals for X, Y and Z direction control. The output for the feedback is obtained from the Signal Access Module (SAM V) box in the form of X, Y, Z and deflection of the probe tip. Using these values, we establish a simple PI control for the X-Y and Z direction movement of the probe. This type of control does not provide better results than the Nanoscope software, but enables us to apply Adaptive Contact Mode (ACM) algorithm as it is only applied on the Z-piezo.

Once we obtain images with the Nanoscope software, we apply external PI control to obtain similar images. We then apply ACM to improvise height tracking of the probe to maintain better contact with the surface with irregularities. We record images with better tracking and can compare with those taken using Nanoscope.

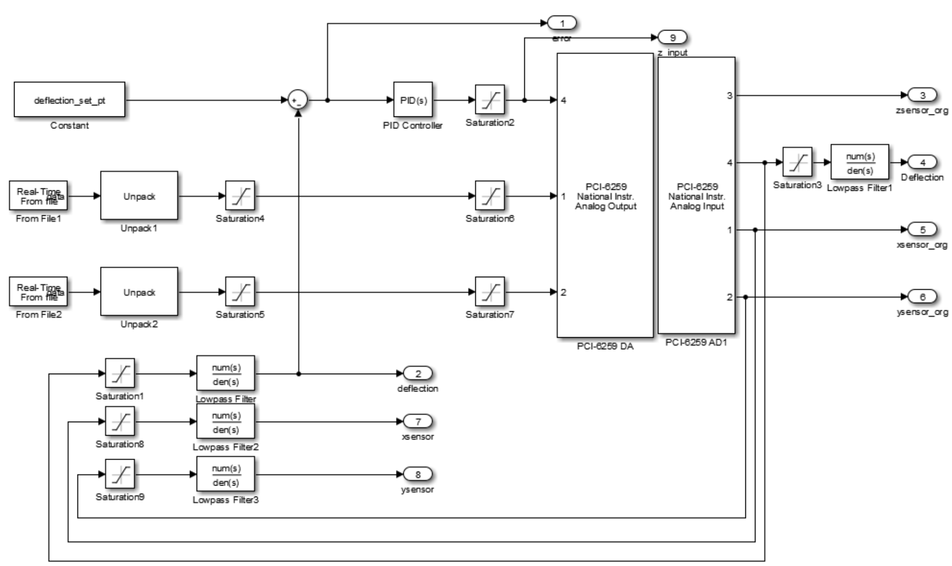

Figure 5.3: PI Control for AFM operation using XPCTarget

For external control, we always apply PI control for the Z-direction movement. For the X and Y movement, we can use different control strategies depending on the scan speed required. PI control is employed at slower speeds (scan rate $\leq$ 10 Hz) as it is inaccurate and unreliable at high speeds. As discussed before, the MIIC technique is used at higher speeds as it has proved to provide accurate results for high speed operation.The main aim of this project is to apply Adaptive Contact Mode Imaging technique and obtain results using PFM. This technique can be applied while using PI Control or MIIC for the X-Y movement. We use both the methods depending on the scan speed.

In case of external control, the Nanoscope software no longer controls the x, y and z direction. The simulink block diagram 5.3 shows external control for the 3 axes stated above. For obtaining PFM signals using Contact Mode imaging, we use the generic lock-in provided by the software to set the reference frequency and bias amplitude. We then apply external control strategies according to the scan speed required.

For the PI control, the block diagram 5.3 needs to bbe modified and a PID Control is applied for the x and y directions.We then set suitable values for the gains and performed Piezoresponse (PR) Imaging. The PI gains are tuned using a standard input before we perform actual imaging. To cover the scan area, we supply a triangle wave for the X direction and a ramp signal for the Y direction. According to the scan size, the scan speed and the resolution required, we create the suitable input for X and Y direction and apply PI control. The combination of both results in a line by line scan a square region on the sample.

For speeds higher than 10 Hz, we use the MIIC technique for x and y direction control. For this, we first need to train the X and Y movement of the probe before engaging on to the sample. This training is done individually and combined (for X and Y) with the same signal inputs as in the PI Control. After performing a iterations, we obtain a good signal which is then fed to the AFM. The Z direction, however, still employs a PI control as stated earlier.

After we perform these experiments we obtain and plot the images for the Height sensor, the deflection, the PR Amplitude and the PR Phase. These images, at varying scan rates, are useful for comparing the 3 approaches for imaging.

Chapter 6

Experimental Results and Conclusion

The experimental results obtained were using the standard Nanoscope software and by using external control. For the nanoscope software, the parameters were set according to the chapter 4. The iterations were done for optimizing the output and then the images were recorded.

Initially, the imaging was done at a low speed of 0.5 Hz. It was gradually increased and the results obtained were analyzed. As the probe and the sample are hard, we can achieve a higher imaging speed without damaging the surfaces. The speed are still limited with respect to the surface roughness of the sample and also the frequency of the voltage applied. Our main aim is to obtain enough information from each pixel as it is scanned by the probe.

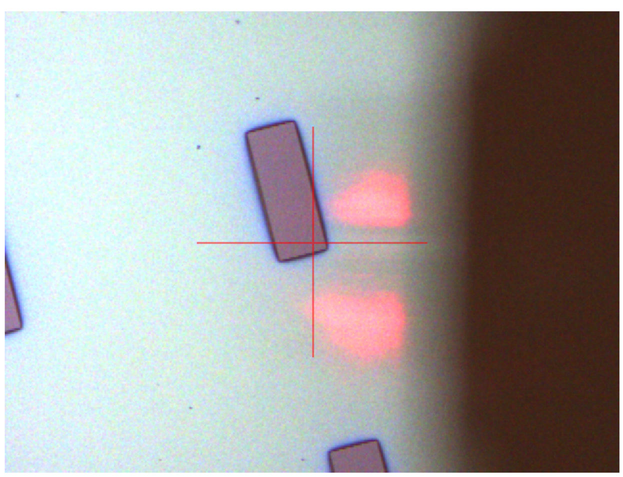

Figure 6.1: View of the Ni-ZnO region of the sample through AFM Optics

We first performed the experiment using the Nanoscope software. We set-up the AFM with the probe and the sample and selected a scan size of 20 μm. The sample used was a patterned Ni-ZnO sample with patterns of the piezo-material on a Silicon dioxide base. This sample was manufactured in such a way that, upon applying voltage across the piezo-region, the polarization alignment was only in one direction. Also, the whole surface contained uniform piezoresponse

The parameters for measuring piezoresponse needed to be adjusted before obtaining a clear image for the PFM amplitude. As there was no phase difference and amplitude change, for obtaining a good contrast in the image, the area selected was half piezo part and half SiO_2 part which did not provide any output.

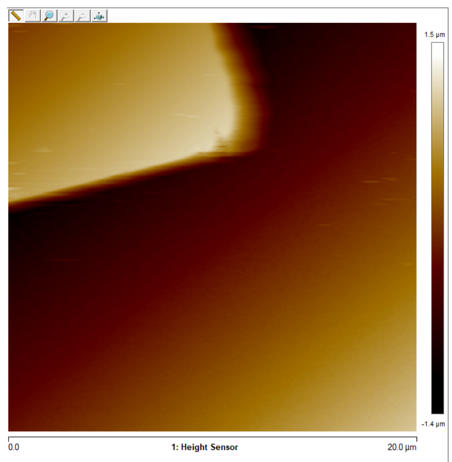

Figure 6.2: Height sensor output

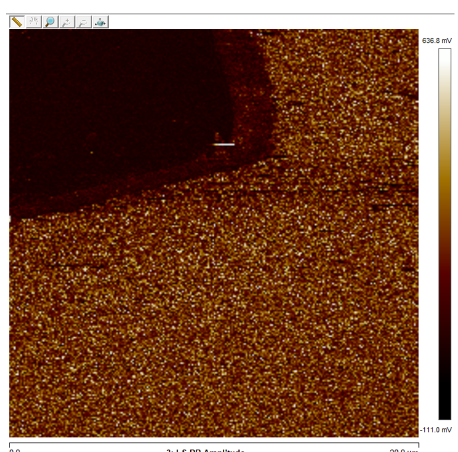

Figure 6.3: Piezoresponse Amplitude

Figure 6.2 shows output of the height sensor of a 20x20 μm part shown in figure 6.1. The top left portion is the piezo-material which has a height of 50 nm. The corresponding Piezoresponse Amplitude is shown in figure 6.3

The following set of experiments were performed with a scan size of 20x20 μm, with increasing levels of scan rates. The AC bias applied was 1000 mV and the frequency for the Lock-In amplifier was set at 1 kHz.

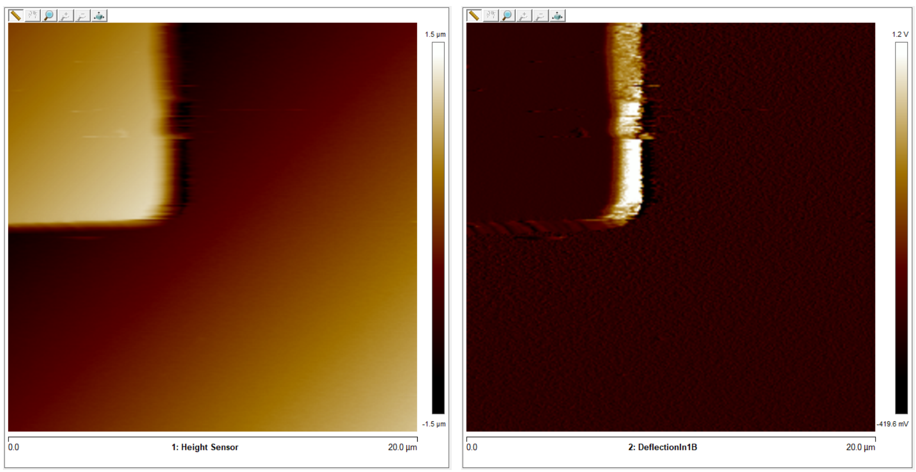

Figure 6.4: Height sensor and Piezoresponse Amplitude with scan rate of 1 Hz

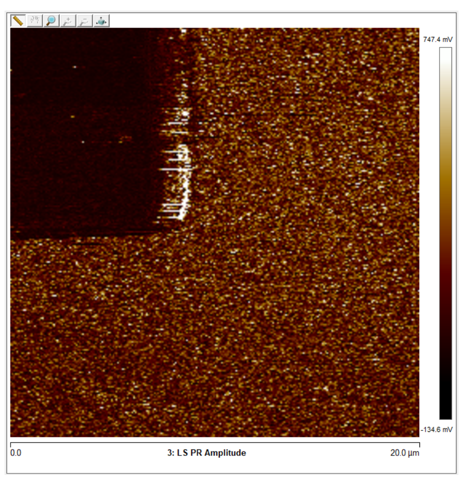

Figure 6.5: Piezoresponse Amplitude with scan rate of 1 Hz

The figure 6.4 shows that the PFM output at the area where the topology changes contains error which can be due to the excess contact or the lack of contact between the tip and the sample.

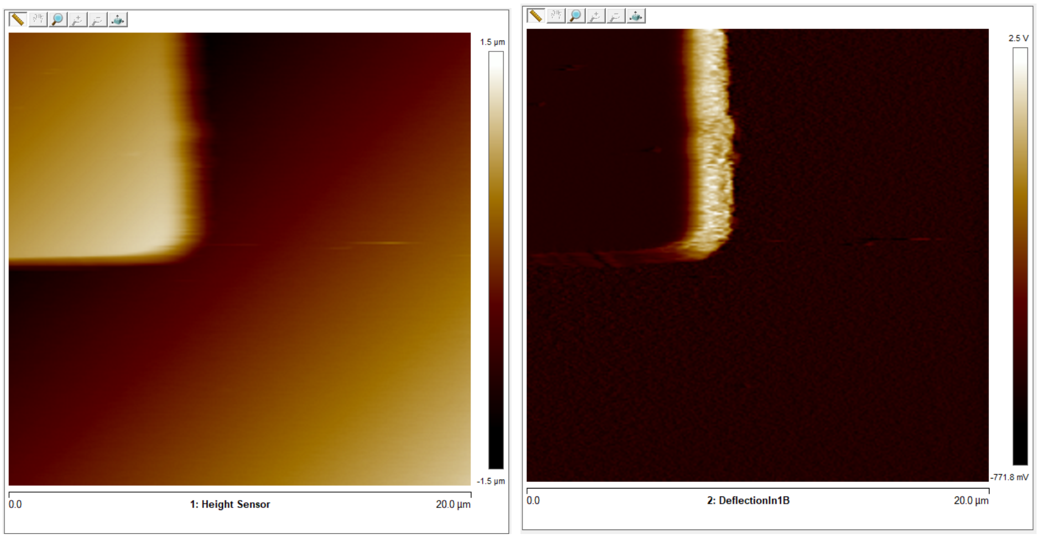

Figure 6.6: Height sensor and deflection signal output with scan rate of 3 Hz

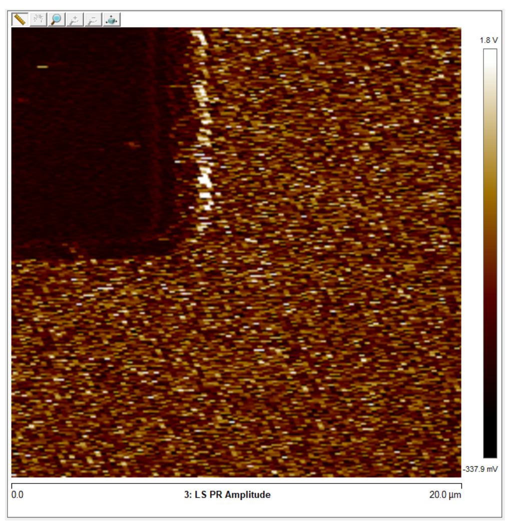

Figure 6.7: Piezoresponse Amplitude with scan rate of 3 Hz

From the images obtained at higher speed, we find that at the area where the topology changes, the error in the PFM signal keeps on increasing which leads to inaccuracies in the data acquired.

Making use of Adaptive Contact Mode technique would ensure proper tip-sample contact and thus provide precise PFM signal even at locations with varying topology. After obtaining images using standard Nanoscope software, we used the XPCTarget

- Matlab package to externally control the probe movement over the surface of the sample. The first step was to use basic PI control for all the three axes of probe movement. The z-direction control can only be done using PI control technique while we can apply different control techniques for x and y direction depending on the scan speed as mentioned earlier. The following images shown are for a scan speed of 5 Hz and using external control. The Adaptive Contact Mode technique was also applied for obtaining images and for comparison purposes.

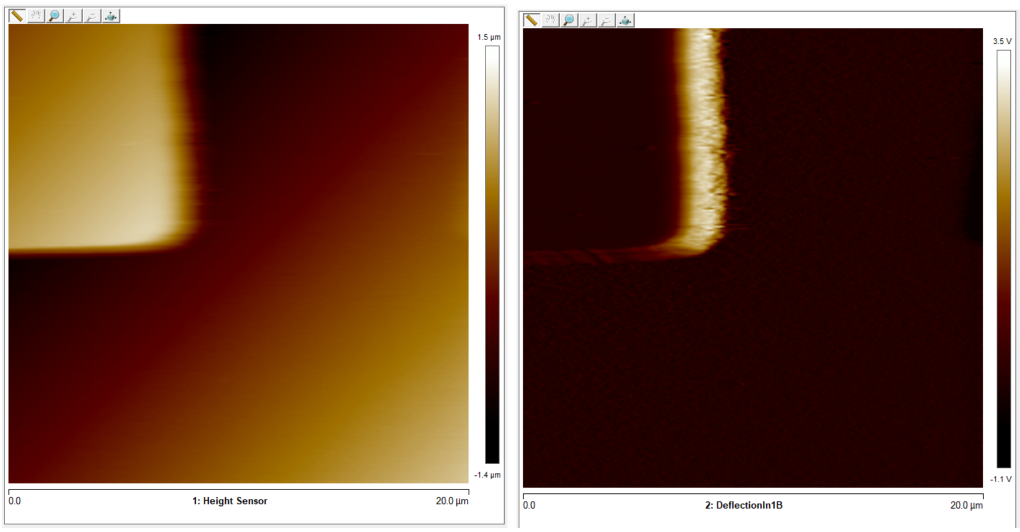

Figure 6.8: Height sensor and deflection signal output with scan rate of 5 Hz

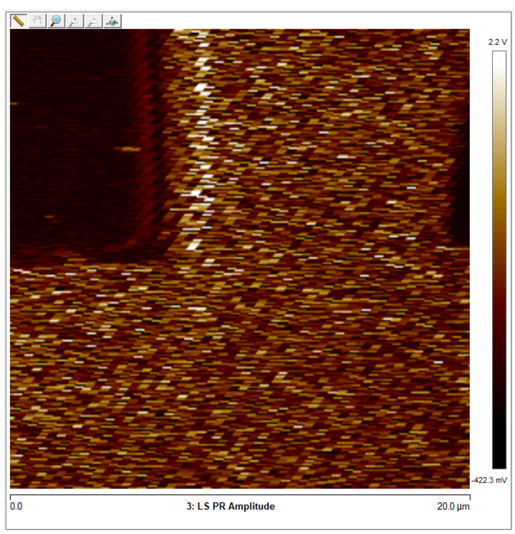

Figure 6.9: Piezoresponse Amplitude with scan rate of 5 Hz

To conclude, we were able to successfully apply external control techniques to obtain

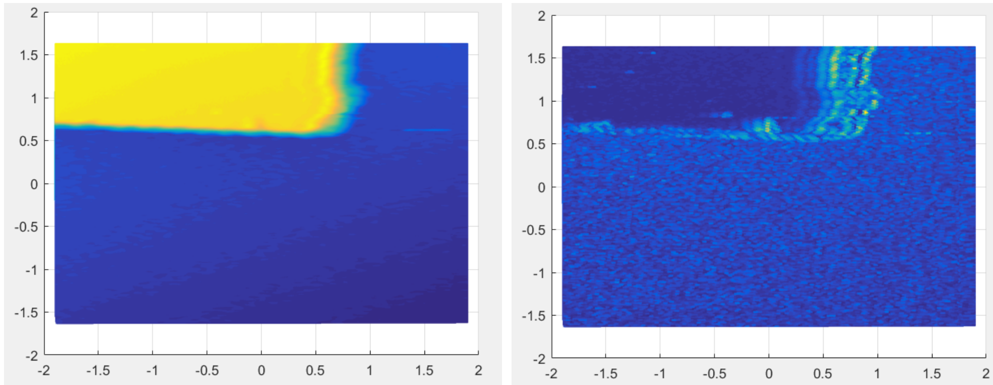

Figure 6.10: Height sensor output (left) and Piezoresponse Amplitudde (right) with scan rate of 5 Hz using external PI Control for all three axes

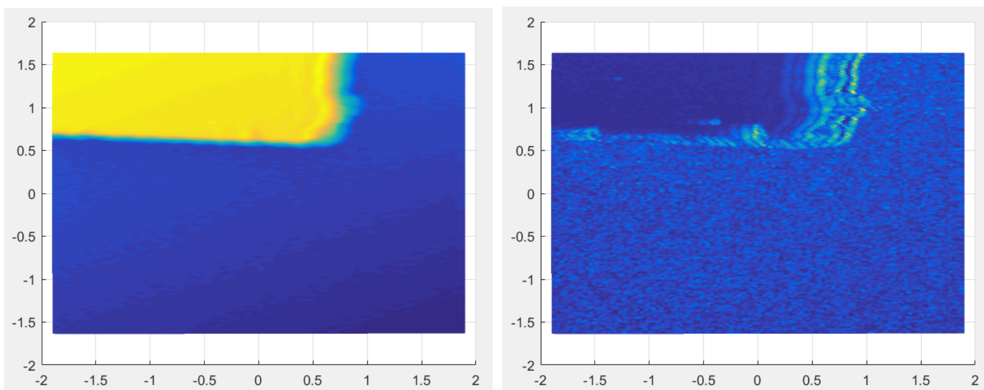

Figure 6.11: Height sensor output (left) and Piezoresponse Amplitudde (right) with scan rate of 5 Hz using external PI Control with Adaptive Contact Mode technique for z-axis

Piezoresponse from the sample. Using such techniques we would be able to increase the scan speed to a much higher amount and with sample with more intricate topographies that the one used for the purpose of these experiments. The idea of contact resonance can also be applied to further improvise the output contrast and can be used along with external control. A better noise reduction technique can also be formulated for compensating the error due to background noise.

Further experiments can be performed at different speeds to prove the effectiveness of Adaptive Contact Mode imaging along with use of other control techniques for x and y direction movement of the probe. We thus conclude that ACM imaging using PF Microscopy yields better results that standard contact mode imaging technique.

Bibliography

Kai-Chih Chang, Yu-Wei Chiang, Chin-Hao Yang, and Je-Wen Liou. Atomic force microscopy in biology and biomedicine. *Tzu Chi Medical Journal*, 24(4):162–169, 2012.

Elisabeth Soergel. Piezoresponse force microscopy (pfm). *Journal of Physics D: Applied Physics*, 44(46):464003, 2011.

Juan Ren and Qingze Zou. High-speed adaptive contact-mode atomic force microscopy imaging with near-minimum-force. *Review of Scientific Instruments*, 85(7):073706, 2014.

A Gruverman and Sergei V Kalinin. Piezoresponse force microscopy and recent advances in nanoscale studies of ferroelectrics. *Journal of materials science*, 41(1): 107–116, 2006.

Ramesh Nath, Ying-Hao Chu, Nicholas A Polomoff, Ramamoorthy Ramesh, and Bryan D Huey. High speed piezoresponse force microscopy:¡ 1 frame per second nanoscale imaging. *Applied Physics Letters*, 93(7):072905, 2008.

YG Kuznetsov, A Low, H Fan, and A McPherson. Atomic force microscopy investigation of wild-type moloney murine leukemia virus particles and virus particles lacking the envelope protein. *Virology*, 323(2):189–196, 2004.

Yu G Kuznetsov, JG Victoria, WE Robinson, and A McPherson. Atomic force microscopy investigation of human immunodeficiency virus (hiv) and hiv-infected lymphocytes. *Journal of virology*, 77(22):11896–11909, 2003.

AJ Malkin, M Plomp, and A McPherson. Application of atomic force microscopy to studies of surface processes in virus crystallization and structural biology. *Acta Crystallographica Section D*, 58(10-1):1617–1621, 2002.

Karsten Rippe, Martin Guthold, Peter H von Hippel, and Carlos Bustamante. Transcriptional activation via dna-looping: visualization of intermediates in the activation pathway of e. coli rna polymerase· $\sigma 54$ holoenzyme by scanning force microscopy1. *Journal of molecular biology*, 270(2):125–138, 1997.

Eric Miller, Tzintzuni Garcia, Scott Hultgren, and Andres F Oberhauser. The mechanical properties of e. coli type 1 pili measured by atomic force microscopy techniques. *Biophysical journal*, 91(10):3848–3856, 2006.

F Braet, C Seynaeve, R De Zanger, and E Wisse. Imaging surface and submembranous structures with the atomic force microscope: a study on living cancer cells, fibroblasts and macrophages. *Journal of microscopy*, 190(Pt 3):328, 1998.

Kyung Sook Kim, Chang Hoon Cho, Eun Kuk Park, Min-Hyung Jung, Kyung-Sik Yoon, and Hun-Kuk Park. Afm-detected apoptotic changes in morphology and biophysical property caused by paclitaxel in ishikawa and hela cells. *PloS one*, 7(1):e30066, 2012.

H-J Butt, P Siedle, K Seifert, K Fendler, T Seeger, E Bamberg, AL Weisenhorn, K Goldie, and A Engel. Scan speed limit in atomic force microscopy. *Journal of microscopy*, 169(1):75–84, 1993.

SR Manalis, SC Minne, and CF Quate. Atomic force microscopy for high speed imaging using cantilevers with an integrated actuator and sensor. *Applied Physics Letters*, 68 (6):871–873, 1996.

ADL Humphris, MJ Miles, and JK Hobbs. A mechanical microscope: High-speed atomic force microscopy. *Applied physics letters*, 86(3):034106, 2005.

Kyong-Soo Kim and Qingze Zou. Model-less inversion-based iterative control for output tracking: piezo actuator example. In *American Control Conference, 2008*, pages 2710–2715. IEEE, 2008.

Sergei V Kalinin, Andrei Rar, and Stephen Jesse. A decade of piezoresponse force microscopy: progress, challenges, and opportunities. *IEEE transactions on ultrasonics, ferroelectrics, and frequency control*, 53(12), 2006.

Dawn A Bonnell, Sergei V Kalinin, AL Kholkin, and A Gruverman. Piezoresponse force microscopy: a window into electromechanical behavior at the nanoscale. *MRS bulletin*, 34(9):648–657, 2009.

P Güthner and K Dransfeld. Local poling of ferroelectric polymers by scanning force microscopy. *Applied Physics Letters*, 61(9):1137–1139, 1992.

Malcolm E Lines and Alastair M Glass. *Principles and applications of ferroelectrics and related materials*. Oxford university press, 2001.

M Labardi, V Likodimos, and M Allegrini. Force-microscopy contrast mechanisms in ferroelectric domain imaging. *Physical Review B*, 61(21):14390, 2000.

Stephen Jesse, Arthur P Baddorf, and Sergei V Kalinin. Dynamic behaviour in piezoresponse force microscopy. *Nanotechnology*, 17(6):1615, 2006.