



Studies of surface interactions at nanometer scale with atomic force microscopy combined with scanning electron microscopy
by Huddee J Ho

A thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy in Physics
Montana State University
© Copyright by Huddee J Ho (1997)

Abstract:

The Atomic Force Microscope (AFM) is an important instrument measuring surface topography and related phenomena; and can study nanometer-scale surface interactions. Surface interactions in ambient air are complicated by surface contamination layers, which do not occur in vacuum or liquid environments. This thesis studies nanometer scale surface interactions, in ambient air, using AFM's high force sensing capability. Several experimental methods were developed and new insights into surface interactions at nanometer scale were obtained. Substantial improvement was made on the lateral resolution of AFM operation in air.

To position the force sensing tip over a specific nanometer scale area, a novel instrument combining AFM and Scanning Electron Microscopy (SEM) was designed, built and commissioned. The system is capable of analyzing AFM tip conditions, in order to study the effects of tip/surface contact. An essentially new method, which monitors the dynamic sensor signal and its fluctuation while changing the tip-sample gap, was developed for vibrating cantilever studies. Sometimes, an electrostatic force occurs in surface contamination, which can be measured with scanning Kelvin probe force microscopy (KFM); this made it possible to develop a technique for probing surface contamination electrical properties.

A capillary force is associated with the contamination layer. Both capillary force magnitude and layer thickness were measured by fitting the approach curve with the capillary force theoretical model. Studying capillary force with cantilevers of differing spring constants, we demonstrated that capillary force can be balanced by a cantilever having a sufficiently large spring constant. With KFM, the contamination layer was found to contain a charge distribution that changes with time. A model is proposed, showing that the surface contamination layer contains a molecular layer bonded tightly to the sample surface. With KFM, dopant concentration was measured on an MBE-grown semiinsulating sample cross-section, and electrical potential scan edge effect was observed. Systematically studying tip-sample contact, two types of contact processes were identified: jump-to-contact and ramp-to-contact, and the conditions under which they occur. A new spatial region (near-contact region), minimizing tip-sample gap without tip-sample contact, was discovered. Operating in the near-contact region is the optimal operating mode of a vibrating cantilever AFM.

STUDIES OF SURFACE INTERACTIONS AT NANOMETER SCALE WITH
ATOMIC FORCE MICROSCOPY COMBINED WITH
SCANNING ELECTRON MICROSCOPY

by

Huddee J Ho

A thesis submitted in partial fulfillment of the
requirements for the degree

of

Doctor of Philosophy

in

Physics

MONTANA STATE UNIVERSITY
Bozeman, Montana

April 1997

D378
H65

APPROVAL

of a thesis submitted by

Huddee J Ho

This thesis has been read by each member of the thesis committee and has been found to be satisfactory regarding content, English usage, format, citations, bibliographic style, and consistency, and is ready for submission to the College of Graduate Studies.

April 15, '97 Lerald J. Lapeyre
Date Chairperson, Graduate Committee

Approval for the Major Department

4-15-97 [Signature]
Date Head, Major Department

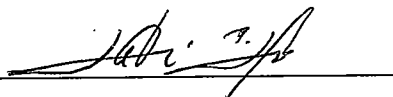
Approval for the College of Graduate Studies

4/16/97 [Signature]
Date Graduate Dean

STATEMENT OF PERMISSION TO USE

In presenting this thesis in partial fulfillment of the requirements for a doctoral degree at Montana State University-Bozeman, I agree that the library shall make it available to borrowers under rules of the Library. I further agree that copying of this thesis is allowable only for scholarly purposes, consistent with "fair use" as prescribed in the U. S. Copyright Law. Requests for extensive copying or reproduction of this thesis should be referred to University Microfilms International, 300 North Zeeb Road, Ann Arbor, Michigan 48106, to whom I have granted "the exclusive right to reproduce and distribute my dissertation in and from microform along with the non-exclusive right to reproduce and distribute my abstract in any format in whole or in part."

Signature



Date

April 14, 1997

To my daughter, Rebecca

ACKNOWLEDGMENTS

This has been the most difficult project of my life. I would like to thank Professor Gerald Jerry Lapeyre whose continued support made this thesis possible.

The research of this thesis was performed at TopoMetrix Corporation's Santa Clara facility, where I received assistance from almost everyone. My greatest thanks to Dr. Paul West. He initiated the idea of doing my Ph.D. thesis in the industry environment, and continuously encouraged me to finish this thesis over the past few years. I am also indebted to other staff at TopoMetrix: Mr. Marc Schuman, who provided electronics support; Ms. Joni Li, whose software tools made my PPM data analysis easy; Dr. Kong Loh, who shared his expertise on loading curves and with whom I had some stimulating discussions on the near-contact mode; and the technical writing experts, Mr. Tony Marek, Mr. Stephen Eichelberger, and Mr. Rick Castagner who helped me on grammar and writing style.

I am also grateful to Dr. James Anderson, who gave me UHV lab training while I was working in the CRISS lab, and contributed many hours of his time in reviewing this dissertation. I would like to thank Professor Jingde Li of Zhongshan University, who taught me all the fundamental skills of building scientific instruments. My four-and-a-half years of research experience on ferroelectric ceramics under his supervision has been extremely helpful for me in designing various PPMs. Thanks are also due to Michael T. Postek of the National Institute of Standards and Technology and Mrs. Nancy Lapeyre for reviewing the thesis and giving a lot of great comments.

I would like to thank my family, for understanding my long work hours. I am deeply indebted to my daughter, Rebecca, who taught me the meaning of life.

TABLE OF CONTENTS

APPROVAL	ii
STATEMENT OF PERMISSION TO USE	iii
ACKNOWLEDGMENTS	v
TABLE OF CONTENTS	vi
LIST OF TABLES	x
LIST OF FIGURES	xi
ABSTRACT	xvii
ACRONYMS	xviii
1. INTRODUCTION	1
2. HISTORICAL REVIEW	7
Introduction	7
Nanometer Scale Force Sensing Techniques	11
Cantilever-Based Force Sensors	12
Tunneling Sensor	15
Capacitance Sensor	15
Interferometer Sensor	15
Optical Deflection Sensor	16
Piezoresist Sensor	16
Shear Force Sensors	17
Impedance Sensor	17
Quartz Tuning Fork Sensor	18
Ranging Techniques for Locating Nanometer Scale Objects	18
Combining STM/AFM and SEM	20
Vibrating Cantilever Mode of AFM Operation	21
Nanometer Scale Surface Electrical Potential Sensors	24
Kelvin-probe Force Microscopy	25
Maxwell Stress Microscopy	27
3. THEORETICAL ANALYSIS	29
Introduction	29
Nanometer Scale Surface Interactions	29

Interaction between Neutral Molecules	30
Interaction between a Sphere and a Flat Surface	33
van der Waals Force	33
Capillary Force	33
Comparing van der Waals Force with Capillary Force	35
Lateral Resolution in Imaging the Electrostatic Properties	36
Mechanical Resonance of a Harmonic Oscillator with Viscous Damping	41
Tip-Sample Contact: Harmonic Oscillator with Non-Damping Force	43
Surface Electrical Potential on Semiconductors	52
Summary	56
4. EXPERIMENTAL APPARATUS	57
Introduction	57
Design of the Combined PPM/SEM System	57
Design Objectives of the Instrument	58
Overview of the Instrument	60
PPM Head	63
Piezoelectric Scanners	65
Sample and Probe Replacement	66
Vibration Isolation System	66
Translation Stages	68
Control System	69
Direct Probe Viewing	69
Testing the Performance of the PPM/SEM System	71
Operation in Vacuum	71
Simultaneous PPM/SEM Operation	73
Aging of Piezoelectric Scanner	74
Vibration Isolation and Atomic Resolution	76
Studies of Tip Conditions	79
Locating Sub-micrometer Features	79
Vibrating Cantilever Mode AFM System	82
System Structure	82
Amplitude Detection Mode	86
Phase Detection Mode	86
System Performance	87
Operation in Vacuum	88
Double-Feedback-Loop Surface Potential Sensing System	92
Potential measurement	92
Spatial Resolution	96
Operation Parameters	99
AFM Probes	99
Silicon Nitride Probe	99
Silicon Probe	100
VLS Silicon Probe	100
Summary	102
5. STUDIES OF AFM LOADING CURVES	103

Introduction	103
Experiments	104
Measuring Force with a Micro Cantilever	104
Measuring a Loading Curve	106
Calibrating a Loading Curve	108
Interpreting a Loading Curve	111
Comparing Loading Curves in Air and in Vacuum	112
Results and Analysis	113
Reproducibility of the Measurements	113
Loading Curves for Cantilevers of Different Spring Constants	117
Surface Contamination Studies	120
Conclusion	125
6. STUDIES OF VIBRATING CANTILEVER MODE	126
Introduction	126
Experiments	128
Experiment One: Changes in Tip Geometry by Periodic Contact	128
Experiment Two: Wear of a Silicon Tip on a Silicon Sample	135
Experiment Three: Surface Modification by Periodic Contact	135
Experiment Four: Monitoring the Tip Approach Process	140
Experiment Five: Comparison of Three Distinct Operation Modes	143
Experiment Six: Estimating Lateral Resolution	149
Results and Analysis	153
Optimal Parameters of VCM Operation	153
Effect of the Spring Constant of Cantilevers	153
Effect of the Driving Frequency	155
Effect of the Vibration Amplitude	157
Amplitude Detection versus Phase Detection	157
Effect of the Phase Setting	157
Working Principle of the VCM Operation	158
Concepts of Contact and Non-Contact	158
Theoretical Model of Tip-Contamination-Sample System	159
Mechanism of Feedback Establishment	164
Mechanism of Tip Damage	165
Classification of the Three Different VCM Operation Modes	168
Examples of Ultra-high Resolution AFM Imaging in Near-Contact Mode	172
Metal Film Samples	172
Semiconductor Samples	172
Insulating Samples	175
Soft Polymer Samples	175
Biological Samples	175
Conclusion	180
7. IMAGING SURFACE ELECTRICAL POTENTIAL	183
Introduction	183
Scanning Kelvin-probe Force Microscopy	183
Imaging the Surface of a PZT Film	186

Designing and Studying a Standard Sample	189
Studies of Sub-Micrometer MBE Grown Layers	197
Sample Preparation	198
Locating the Area of MBE Layer	200
Surface Potential Imaging	200
Results and Analysis	203
Conclusion	205
8. CONCLUSIONS	207
Proximal Probe Microscopy	207
Experimental Apparatus and Methods	208
Surface Contamination and Capillary Force	209
Optimal Operation Conditions for the Vibrating Cantilever Mode AFM	210
Nanometer Scale Surface Potential Measurement	211
Future Studies	212
REFERENCES CITED	213

LIST OF TABLES

Table	Page
1. Parameters of the three silicon cantilevers used for the loading curve comparison.....	118
2. Experimental parameters of the three different VC operation modes.....	143
3. Comparison of energy dissipation.....	168
4. Characteristics of the three VCM operation.....	171

LIST OF FIGURES

Figure	Page
2.1. The operation of a PPM. (a) Initial set-up; (b) The tip approach process; (c) The scanning process.....	9
2.2. Cantilever-based force sensor.....	13
2.3. Shear force sensor.....	14
2.4. The working principle of the AFM vibrating cantilever mode (VCM).....	22
3.1. The force created by the Lennard-Jones Potential.....	32
3.2. The capillary attraction between a sphere and a flat surface.....	34
3.3. Comparison of the van der Waals force and capillary force between a sphere and a flat surface for two different values of the radius, R	35
3.4. Diagram of a single charge and a conducting sphere.	36
3.5. Definition of the lateral resolution.	38
3.6. Electrostatic force between a single charge and a conducting tip.....	40
3.7. The resonance spectra of a driven damped harmonic oscillator.	42
3.8. Jump-to-contact and ramp-to-contact.	44
3.9. A simplified model of a vibrating probe in an attractive interaction with the sample. The interaction force has a positive gradient.....	46
3.10. The total force applied to the AFM probe and the conditions of JTC: (a) JTC occurs when $x < x_J$; (b) JTC never occurs; (c) JTC always occurs; (d) JTC occurs only for $x_J = 0$	48
3.11. Conditions of JTC as a function of the relative tip-sample gap μ and the relative spring constant λ	49
3.12. Simple Harmonic Oscillator under an attractive force between the tip and the sample. In this example, $x_0 = 50$ nm, $x_1 = 10$ nm.	51

3.13. Energy loss when the tip encounters a rigid surface in RTC.	53
3.14. (a) Diagram of a sharp p-n junction; ⁵⁵ (b) Dopant concentration and net charge density; (c) Electrical potential.	55
4.1. Configuration diagram of the PPM/SEM combination.	59
4.2. Diagram of the PPM integrated with a scanning electron microscope.	61
4.3. Picture of the PPM unit removed from the SEM specimen chamber.	62
4.4. Diagram of the PPM head. (A) laser diode, (B) primary deflection mirror, (C) secondary mirror, (D) photoelectric sensor, (E) Sample, (F) piezo tube, (G) AFM scanner, (H) STM scanner, (I) primary XY translator, (J) manual adjustment screw, (K) motorized screw, (L) DC motor.	64
4.5. The vibration isolation system.	67
4.6. Schematic diagram of the electronic control system.	70
4.7. Examples of different tip conditions of a type of commercially available silicon AFM tip. (a) A tip that was not etched completely. (b) A regular sharp tip.	72
4.8. Images of highly-oriented-poly-graphite (HOPG) acquired in the SEM. (a) AFM force image. The field of view is 30 Å. (b) STM tunneling current image. The field of view is 12 Å. The bias voltage was 50 mV. The tunneling current was 1 nA.	77
4.9. The background noise spectra of the feedback signals.	78
4.10. SEM micrograph of a VLS silicon AFM tip before (a) and after (b) usage. The image on the right side is the enlarged tip area of the left image. The increase of the magnification is ten times. The scale bar on the bottom is for the image on the left side. For the right image, each division represents 66.7 nm.	80
4.11. Near-contact AFM image of an aluminum grain sample. There are two deep scratch marks and one shallow scratch mark in the image area. The image size is 1.6 mm × 1.6 mm. The image was acquired in air. The cantilever vibration amplitude was 2 nm.	81
4.12. Studies of micro crack in ceramic. (a-c) SEM images; (d) an AFM image. The size of the AFM topographic image is 2 × 2 μm.	83

4.13. Diagram of the electronics for operating the vibrating cantilever mode AFM.....	84
4.14. Selecting the operation frequency.....	85
4.15. Transfer function of the control electronics in amplitude mode.....	89
4.16. Frequency spectrum of amplitude and phase signals.....	90
4.17. Selecting the drive frequency for vacuum operation.....	91
4.18. Vibrating cantilever AFM images acquired in two environments: (a) in air; (b) in vacuum. The sample is an aluminum film on silicon substrate.....	93
4.19. Diagram of the double-feedback loop surface potential sensing system.....	94
4.20. Wiring diagram of the surface potential imaging system.....	95
4.21. Testing the potential measurement.....	97
4.22. Spatial resolution of KFM.....	98
4.23. The AFM probes used in this dissertation. (a) Silicon nitride with FIB sharpening; (b) Single-crystal silicon probe; (c) Silicon tip grown with the VLS technique.....	101
5.1. Diagram of the cantilever bending.....	105
5.2. Mechanism of force detection with the optical deflection force sensing system.....	106
5.3. An experimental loading curve.....	107
5.4. An example of loading-curve calibration using an approach curve measured in air: (a) As measured. (b) Cantilever-deflection corrected from the measured curve in (a).....	110
5.5. Jump-to-contact and adhesive pullback.....	112
5.6. Repeatable loading curves measured consecutively.....	114
5.7. Loading curves measured on different samples.....	115
5.8. Loading curves for cantilevers of different spring constant.....	119
5.9. Comparison of the approach curves in the loading curves of three cantilevers with different spring constants near the tip-surface contact point.....	121
5.10. The loading curves measured in air and in vacuum with the same cantilever on the same sample area.....	123
5.11. Fitting the approach curve with a simplified model of capillary attraction force. $R = 800$ nm, $G_0 = 11.6$ nm.....	124

6.1. The resonance spectrum of the VLS silicon probe.....	129
6.2 SEM image of the VLS silicon tip. (a) Before any topographic AFM image was acquired, (b) after three hours of imaging with a small (8 nm) vibration amplitude with a 40% reduction in amplitude, and (c) after one topographic image with 80 nm vibration with a 10% reduction in amplitude. The scale dots on the lower right corner is for the image in the left. The right image corresponds to the box in the left image. Its scale is 1/10 of the left image.....	131
6.3. Topographic images (shaded view) of aluminum grains: (a) with 8 nm vibration amplitude and (b) with 80 nm vibration amplitude. The size of both images is 800×800 nm.	132
6.4. Spatial frequency spectrum of the topographic images illustrated in Figure 6.3.	134
6.5. SEM image of a silicon tip before imaging a silicon surface (a) and after imaging with an vibration amplitude of 100 nm and a 10% reduction in amplitude (b).	136
6.6. The mechanism of sample damage.....	138
6.7. The near-contact AFM images (a)-(c) show that the sample is damaged by periodic contact operation.	139
6.8. The SEM micrographs taken before and after the experiment (shown in Figure 6.7) show no obvious change in the tip profile.....	141
6.9. The transition from traditional non-contact to near-contact.	142
6.10. Resonance spectrum of the silicon tip (Model 1650-00 #25).	144
6.11. Topographic images of a polished alumina-silicate glass surface with three distinct operation modes. (a) Near-contact mode; (b) Traditional non-contact mode; (c) Periodic contact mode. The image size is 100 nm × 100 nm.	145
6.12. Line profiles of the same features in the three VC modes.	147
6.13. Spatial frequency spectrum of the three VC mode operation conditions.	148
6.14. SEM images of the tip before and after acquiring the three VC mode AFM images.	150

6.15. Topographic image of a niobium film sample that has the potential of being a standard sample for tip geometry characterization. (a) The image is displayed with pseudo-2D color (90/140); (b) the area of the box in (a), displayed in 1D color.....	151
6.16. A set of consecutive line profiles from a zoomed section of the image (Figure 6.15(b)). The scan lines are displayed by shifting 1 nm upward from the previous scan line.	152
6.17. Relationship between the drive frequency and the feedback setpoint.....	156
6.18. Microscopic model of tip-contamination-sample system.....	161
6.19. Molecular model of tip approaching process in ambient air.....	163
6.20. Diagrams of the mechanism of the three VC mode operation conditions: (a) traditional non-contact condition; (b) periodic contact condition; (c) near-contact condition.....	166
6.21. Classification of three VC modes by the three different stages of tip-sample interaction.....	170
6.22. Aluminum film on a silicon wafer.....	173
6.23. Atomically flat cleavage surface of GaAs.	174
6.24. Polished alumina-silicate glass substrate.....	176
6.25. Epoxy acrylic coating.	177
6.26. Two types of soft polymer thin-film samples.	178
6.27. Human infant hair sample.	179
7.1. Separating the surface electrostatic information from the topographic information. (a) There is only the non-contact topographic feedback loop. The sample has no surface potential change. (b) There is localized surface charge on the sample. (c) Topographic feedback plus the potential feedback for the sample with localized surface charge.....	185
7.2. Images of a PZT ceramic surface: (a) before, (b and c) after the surface potential feedback loop is turned on.	187
7.3. Comparison of topography line profiles before and after the surface potential feedback loop is turned on.	188
7.4. Time dependence studies of KFM on a PZT ceramic surface.....	190

7.5. Design of the standard sample for electrical potential measurement.	191
7.6. Simultaneous topographic (a) and surface potential (b) images of the standard sample. The electrodes labeled "A" were always connected to ground. The electrodes labeled "B" were connected to a voltage source controlled by the software. Each image has 300 scan lines. Each line has 300 data points.	192
7.7. Line No. 80 of the images showed in Figure 7.6.	194
7.8. Three line profiles from the potential image show the relative of the potential changes.	195
7.9. The GaAs MBE grown multilayer sample.	198
7.10. The surface topography of cleavage surfaces. (a) a bad cleave. (b) a good cleave.	199
7.11. SEM view showing positioning of the AFM tip over the area of MBE layers.	201
7.12. Simultaneous topographic and surface potential images of the MBE layers.	202
7.13. A line profile taken from the surface potential image. (a) The location of the line in the KFM image; (b) Comparing the measured data with the calculated data.	204
8.1. Classification of various SFM modes.	208

ABSTRACT

The Atomic Force Microscope (AFM) is an important instrument measuring surface topography and related phenomena; and can study nanometer-scale surface interactions. Surface interactions in ambient air are complicated by surface contamination layers, which do not occur in vacuum or liquid environments. This thesis studies nanometer scale surface interactions, in ambient air, using AFM's high force sensing capability. Several experimental methods were developed and new insights into surface interactions at nanometer scale were obtained. Substantial improvement was made on the lateral resolution of AFM operation in air.

To position the force sensing tip over a specific nanometer scale area, a novel instrument combining AFM and Scanning Electron Microscopy (SEM) was designed, built and commissioned. The system is capable of analyzing AFM tip conditions, in order to study the effects of tip/surface contact. An essentially new method, which monitors the dynamic sensor signal and its fluctuation while changing the tip-sample gap, was developed for vibrating cantilever studies. Sometimes, an electrostatic force occurs in surface contamination, which can be measured with scanning Kelvin-probe force microscopy (KFM); this made it possible to develop a technique for probing surface contamination electrical properties.

A capillary force is associated with the contamination layer. Both capillary force magnitude and layer thickness were measured by fitting the approach curve with the capillary force theoretical model. Studying capillary force with cantilevers of differing spring constants, we demonstrated that capillary force can be balanced by a cantilever having a sufficiently large spring constant. With KFM, the contamination layer was found to contain a charge distribution that changes with time. A model is proposed, showing that the surface contamination layer contains a molecular layer bonded tightly to the sample surface. With KFM, dopant concentration was measured on an MBE-grown semiinsulating sample cross-section, and electrical potential scan edge effect was observed. Systematically studying tip-sample contact, two types of contact processes were identified: jump-to-contact and ramp-to-contact, and the conditions under which they occur. A new spatial region (near-contact region), minimizing tip-sample gap without tip-sample contact, was discovered. Operating in the near-contact region is the optimal operating mode of a vibrating cantilever AFM.

ACRONYMS

AFM	atomic force microscope/microscopy
EFM	electrical force microscopy
FIB	focused ion beam
JTC	jump-to-contact
KFM	Kelvin-probe force microscope/microscopy
MBE	molecular beam epitaxy
MFM	magnetic force microscopy
NSOM	near-field scanning optical microscope/microscopy
PPM	proximal probe microscope/microscopy
PZT	lead-zirconium-titanate
RHEED	reflection high energy electron diffraction
RTC	ramp-to-contact
SEM	scanning electron microscope/microscopy
SFM	scanning force microscope/microscopy
SPM	scanning probe microscope/microscopy
STM	scanning tunneling microscope/microscopy
UHV	ultra high vacuum
VCM	vibrating cantilever mode
VLS	vapor-liquid-solid
VSM	vibrating sample mode

CHAPTER 1

INTRODUCTION

Atomic Force Microscopy (AFM) is a new form of microscopy for imaging surface topography in the nanometer scale. The method was first introduced by IBM and Stanford scientists in 1986,¹ where they demonstrated the proof-of-concept and the outline of the various operation modes. Considerable effort was made in the development and application of this kind of instrument. Perhaps the first method commonly used was to drag the tip across the surface and measure the deflection of the tip mounted on the end of a cantilever.² Although significant success was achieved with this method,^{3,4} it can result in significant tip and/or sample damage.⁵ To obtain non-destructive measurements, tip-sample contact needed to be minimized, which was accomplished by using vibrating cantilever methods.⁶

To understand the mechanism of the microscopy techniques, one should study the tip-sample interaction, which is influenced by the environment. For air operation, the interaction is complicated by the existence of surface contamination. The commonly used technique to study tip-sample interaction is to measure the dependence of the cantilever force on the tip-sample gap.⁷ As frequently observed in loading curves measured in air, the tip can be pulled into contact with the sample by the sample's surface and its contamination. This jump-to-contact behavior results in tip and/or sample damage.

Two major operating modes were developed for the vibrating cantilever AFM. These old modes presented a number of problems when operating in ambient air. In the traditional non-contact mode,⁶ the existence of surface contamination layer results in a large tip-sample gap, which

limits AFM's lateral resolution. In microscopy, lateral resolution generally means the smallest feature that can be resolved by a microscope. When the tip is brought into the contamination layer, the capillary force between the tip and the sample can capture the tip to the sample surface, which stops the vibration of the cantilever. In the "tapping" mode⁸ proposed later, the cantilever is set to vibrate at much larger amplitudes (> 20 nm), which results in periodic contact between the tip and the sample. Such physical contact, although it can be very light, damages the tip and/or sample when operating in air, and limits the lateral resolution.

The objective of this thesis is to understand the physics of nanometer scale surface interaction with the presence of surface contamination layers, and to understand the working mechanism of vibrating cantilever mode AFM to optimize imaging resolution. The results of this thesis research can be summarized in five areas. The first is the development of new experimental apparatus and methods. The second is the understanding of the surface contamination layers in ambient air. The third is the understanding of the capillary force due to the surface contamination at nanometer scale. The fourth is the study of surface electrical potential at nanometer scale. The fifth is the understanding of surface contact at nanometer scale in ambient air. The observation and discoveries on the physics of nanometer scale tip-sample interaction in ambient air leads to new insights on the working mechanism of vibrating cantilever AFM, and the development of a new operating mode for the vibrating cantilever AFM in air.

To study the nanometer scale surface interaction, we developed a number of new experimental techniques during the course of this thesis work. To precisely position the force sensing tip over a specific nanometer scale area, we built the first instrument that combines AFM and scanning electron microscopy (SEM). This system is also capable of analyzing the conditions of the force-sensing tip for studying the effects of tip-surface interactions. To permit proper comparison between conventional loading curve measurements and theoretical models of surface interactions, an analysis method for removing the effect of cantilever deflection for the loading curves was used. A

novel method was demonstrated for studying the interaction between a vibrating tip and a surface, in which the dynamic sensor signal and its fluctuations are monitored while changing the gap between the tip and the surface. To study the surface electrostatic interaction, a scanning Kelvin-probe force microscopy (KFM) system was built as an addition to the vibrating cantilever AFM. We developed a new method that can detect the surface contamination layer by monitoring the time dependence of the KFM signal.

It is generally acknowledged that a "contamination" layer exists on the surface of a sample in ambient air.⁹ However, the nature of this contamination layer is not well understood. In this thesis, the thickness of the contamination layer was measured for the first time by comparing the loading curves measured in air and in vacuum and fitting the approach curve in air with the theoretical model of capillary force. By comparing the loading curves measured in air and under vacuum, we found that the contamination layer is a major factor contributing to the fluctuation of loading curve measurements. With the KFM, we found that the contamination layer contains a space charge distribution that varies with time, which confirms the liquid-like property of the contamination. We proposed a structural model of the surface contamination layer that consists of a molecular layer that is bonded tightly to the sample surface. This model explains our observations of various surface interactions at nanometer scale.

It is generally acknowledged that when a tip and a surface make contact in the presence of contamination layers, a capillary force exists between the tip and the surface.¹⁰ The magnitude of the capillary force between a nanometer sized tip and a surface coated with contamination layers is not well understood. It was believed that an AFM cantilever could not overcome the capillary force and would be captured to the surface when the tip-sample gap is less than a few nanometers. In this thesis, we measure the magnitude of the capillary force by fitting the theoretical model of capillary interaction to the loading curves that we measured. By studying the capillary force with cantilevers of different spring constants, we demonstrate that the capillary force can be balanced by cantilevers having adequately large spring constants.

Using KFM, the time dependence of the electrostatic interaction after a step change of the bias voltage was investigated. The process of redistribution of surface charge was observed. The surface electrical potential on a cross section of an MBE grown semi-insulating sample was measured and fitted with a theoretical model to obtain the dopant concentration. The unique edge-effect phenomenon of electrical potential was observed at the edge of the cross-section of an MBE prepared sample.

It is generally acknowledged that the tip and/or sample may sustain damage when they are brought into contact. However, the physics of nanometer scale surface contact in the presence of surface contamination layers, and the mechanism of surface damage are not well understood. With the combined AFM/SEM system, we found three types of nanometer scale tip damage. We also observed, for the first time, the sample damage due to minimal contact when the sample is scanned with large-amplitude vibrating cantilever AFM in ambient air. Two types of contact processes were identified: jump-to-contact (JTC) and ramp-to-contact (RTC). We established the conditions in which JTC and RTC occur. We then discovered a very narrow spatial region at close proximity to the surfaces, designated as the "near-contact region", where the tip-sample gap is minimized and physical contact between tip and sample does not occur. Maintaining an AFM tip in the near-contact region is critical for obtaining maximum lateral resolution in AFM images. We established the optimal parameters in vibrating cantilever modes for making high resolution AFM topographic images.

The suggestion of completing this Ph.D. thesis research at TopoMetrix was initiated by Dr. P. West in early 1992 and approved by the Graduate Committee in February 1992. The design of the first version of the combined PPM/SEM instrument began on January 1993. The design of the final version was completed on August of 1993 and was granted a United States Patent on October 1995.¹¹ Most of the experimental work was done in 1994 and the first half of 1995. Some of the results were presented at the 1995 APS March Meeting, and a paper was submitted to *Scanning*

for publication on September of 1995 and published in August 1996.¹² The writing of this thesis began in the late summer of 1995 and was severely interrupted by the decease of my daughter, Rebecca, at the end of 1995. The review of the thesis by the advisors began on August of 1996, and the defense was on March 7, 1996. In addition to the academic results, this thesis research also generates three major results of interest to the PPM industry. It resulted in the world's first and still the only commercial SPM instrument combined with a SEM, the first commercial scanning electrical potential microscopy, and a new working mode, near-contact mode, for vibrating cantilever AFM.

Most of the work in this thesis was done by the author alone, including the design and construction of the instrument, planning and set-up of the experiments, processing and analysis of data, and development of theoretical models and analysis. During the course of this thesis work, direct assistance was given by a number of people. Dr. Paul West initiated the original concept of a combined AFM/SEM instrument and provided the name, Near Contact, to our new AFM operating mode. Mr. Marc Schuman designed the electronics for the double-feedback-loop system. Mr. E. Hazaki of Hitachi Scientific Instrument provided the first evidence of the laser-heating problem in the combined PPM/SEM instrument, which was later corrected by the author's modification of the design. Fabrication of the KFM standard sample was arranged by Dr. P. Chiang. The MBE-grown multilayer GaAs sample was specially grown by Dr. H.-S. Wu for this thesis.

The writing of this thesis has been a significant learning process in my career. I learned how to communicate professional knowledge with people who come from different backgrounds, and acquired the skills associated with drawing conclusions from vast amounts of data, ideas, and conjectures.

Since PPM is a relatively new area of scientific study, a large number of new concepts may be encountered by the reader. The layout of this thesis is presented in such a way as to help the reader understand the body of the thesis work. Chapter 2, Historical Review, presents the history of pre-

vious developments in PPM related areas. It also introduces a number of important concepts that will be used in discussion of the following chapters. Chapter 3, Theoretical Analysis, presents the theoretical analysis of various subjects related to this thesis work. It focuses on the discussion of nanometer scale tip-sample interaction. Chapter 4, Experimental Apparatus, describes the instruments and experimental methods used in this thesis work. Chapter 5, Studies of AFM Loading Curves, discusses the experiments and results on the study of the nanometer surface interaction with a DC method (loading curve measurement). Chapter 6, Studies of Vibrating Cantilever Mode (VCM), discusses the experiments and results on the study of the nanometer surface interaction with an AC method (vibrating cantilever). It also presents a theoretical model that explains the nanometer scale tip-sample interaction in air and the working mechanism of VCM, and the new operation mode that maximizes lateral resolution for AFM in air. Chapter 7, Imaging Surface Electrical Potential, deals with the experiments with KFM. Chapter 8, Conclusion, presents the summary of the observations and discoveries of the physics of nanometer scale surface interaction in air and the working mechanism of AFM, and the development of experimental techniques that resulted from this thesis research.

CHAPTER 2

HISTORICAL REVIEW

Introduction

Using a material probe to detect a surface by touching it to the specimen is a natural concept. For instruments that measure surface topography, a material stylus maintained at close proximity to the sample surface is used to perform the measurement. For many years, surface profilers with a stylus tip have been used to measure the surface profile.¹³ The topografiner,¹⁴ invented in 1972, was the first surface profiler that achieved sub-micrometer lateral resolution. It was the first instrument using piezoelectric ceramics to scan the tip across the surface, and it achieved one nanometer vertical resolution with a feedback system. About ten years later, a breakthrough was made by G. Binnig and H. Rohr¹⁵. They demonstrated that the atomic structure on surfaces could be imaged using quantum tunneling between a sharp metal tip and the sample. This work opened the door to using material probes as a microscopy tool. They called their new instrument the Scanning Tunneling Microscope (STM).

Although it is powerful enough to image surface atoms, the STM has a significant shortcoming. Since the electron tunneling effect is used in an STM to detect the sample surface, it requires the sample to have some conductivity. However, a large portion of the material world is non-conducting, which substantially limits the application of the STM. To image the topography of non-conducting materials, a tip-sample interaction independent of sample conductivity must be used. The interaction forces that exist between a tip and any solid sample are substantially large when the tip is brought to within a few nanometers of the sample surface. These forces exist inde-

pendently of the electrical conductivity of the sample. Therefore, a force sensor that detects these forces can be used to sense the proximity of any sample surface. In 1985, a high-sensitivity force sensor was developed based on an STM. With this force-sensing technique, the first atomic force microscope (AFM) was invented.¹

The STM, AFM, and all the related technologies that use a material probe to perform microscopic imaging at close proximity of a sample surface have been grouped into the term scanning probe microscopy (SPM). Recently, a new term, Proximal Probe Microscopy¹⁶ (PPM), has been used. This new term is more appropriate in distinguishing solid material probes from particle probes, such as electron probes and ion probes. The word "proximal" emphasizes that our scanning probes are operating in the near-field¹⁷ condition.

Each PPM involves a physical property that strongly depends on the spacing between the tip and the sample, and a sensing system built to detect this physical property. The tip-sample gap is controlled by a piezoelectric actuator, and this spacing can be maintained by feedback electronics that receives the input signal from the sensing system and controls the piezoelectric actuator. Because this piezoelectric actuator moves the probe perpendicular to the sample, it is commonly called the *Z piezo*. When the tip-sample gap is maintained constant by the feedback electronics, we say the PPM system is *in topographic feedback* (or *in feedback* as the abbreviation), and call the system a *topographic feedback loop*. While the tip-sample gap is maintained constant, that is, when the PPM is in feedback, the surface topography can be obtained from the displacement of the *Z piezo*, which is a function of the voltage applied to the piezo. A piezoelectric XY scanner (also called *XY piezo*) is another important component of PPM, which scans the tip across the sample surface while the *Z piezo* voltage is being recorded. The spatial distribution of the recorded *Z piezo* voltage can be used to generate an image representing the topography of the sample being measured, which is also called *to image* the surface topography. The smallest step size of a piezoelectric scanner can be less than one angstrom, which helps the PPM to image atomic structures.

The operation of PPM involves two major steps (Figure 2.1). The first is tip-approach, which

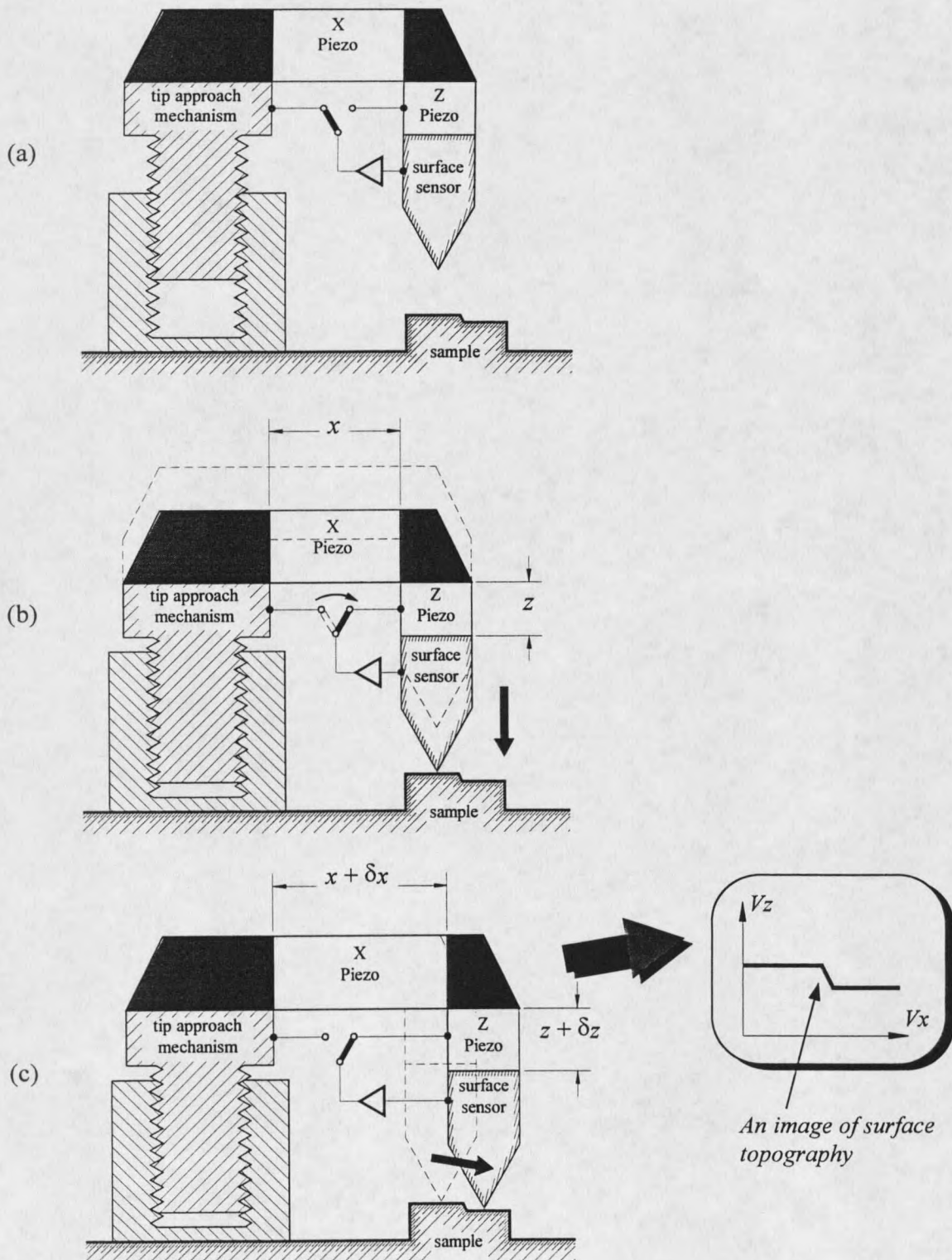


Figure 2.1. The operation of a PPM. (a) Initial set-up; (b) The tip approach process; (c) The scanning process.

is a process that brings the PPM probe from a macroscopic tip-sample gap to where the tip-sample gap is on the order of a few nanometers or less. Compared to other scanning microscopies, this step is unique for proximal probe microscopy. The second step is to scan the probe across the sample surface to acquire an image while the tip is maintained in the proximity of the sample surface by the topographic feedback loop. The topographic image is generated by plotting the Z piezo voltage versus the XY piezo voltages.¹⁸ Since a topographic image of PPM is created by the voltages applied to the piezo scanners, the image is generally independent of the physical property used to sense the sample surface.

By the use of additional sensors or additional sensing techniques, other surface physical properties, such as electrical potential, can also be measured simultaneously with topography. While the XY piezo scans the tip across the sample surface, these other physical properties can be recorded along with the topography. The spatial distribution of a recorded signal output of an additional sensor can be used to generate an image representing the physical property being measured, which is also called *to image* this physical property.

PPM has a number of advantages over other microscopy techniques. Its resolution is higher than traditional optical microscopy and the SEM, permitting it to image local surface topography and electrical structure at the atomic scale. Many other physical and chemical properties can also be imaged in sub-micrometer scale resolution, and experiments on the nanometer scale is possible. All PPMs can be operated in air, vacuum, or other environments, which greatly increase the application area of the PPM.

The topographic images that PPM generates are three-dimensional. On very flat samples (roughness < 10 nm), PPM images have much higher topography contrast than Scanning Electron Microscopy (SEM). Because the contrast of a SEM image is created by the difference in the secondary emission of surface material or change of slope on surface topography, a flat surface of uniform material results in very low contrast for SEM. However, PPM records surface topography

with the position of the piezoelectric actuator that controls the feedback. The image contrast in surface topography is limited by the voltage noise on the piezoelectric actuator and environmental vibration, which can usually be made small enough to allow the system to detect topographic variation of less than one nanometer. For SEM, non-conductive samples must be coated with conductive material to obtain high-resolution images. For AFM imaging, since the image mechanism does not require any conductivity of the sample, the conductive coating is not necessary.

The major limitations of PPM are caused by the limited Z piezo range (usually smaller than 10 μm) and the tip geometry. PPM imaging is restricted to relatively flat samples. The fluctuation of surface topography must be smaller than the range of Z piezo. However, SEM does not have such a restriction, enabling imaging of porous or very rough sample.

Because of the prospect of imaging atoms, the novel imaging mechanisms, capability of exploring the nano world, along with modern computer technologies, and the quest by researchers from various fields to see ever smaller objects, PPM quickly became one of the most important microscopy techniques for academic research and industrial applications.

Nanometer Scale Force Sensing Techniques

To perform high-resolution imaging, the tip must be brought very close to the surface, usually 10 nm or less. A sensing technique that detects the change in tip-sample gap is necessary to bring the tip into proper working position. Since interaction forces between the tip and the sample strongly depend on the tip-sample gap, various surface-force-sensing techniques were developed for controlling the tip-sample gap. A PPM equipped with one of the surface-force sensing techniques is called a Scanning Force Microscope (SFM). Depending on the direction of the force to be detected (vertical or lateral), the microscopic force sensing techniques can be classified into two types: *normal force sensor* and *shear force sensor*.

Most of the microscopic force sensing techniques used in the SFM are normal force sensors which include a microscopic cantilever that converts the variation of the tip-sample interaction

force into the cantilever movement, and a displacement-sensing device that detects the movement of the cantilever (see Figure 2.2). Historically, this type of microscope has been called the Atomic Force Microscope (AFM).^{1,19}

The shear-force sensing technique uses a rigid tip (like in the STM) which is positioned perpendicular to the sample surface, and is set to vibrate parallel to the surface (see Figure 2.3). When the gap between the tip and the sample is small enough, the tip-sample interaction will be strong enough to change the resonance characteristics of the tip, which can be detected. The detailed mechanism of the change of the resonance characteristics will be explained later in Chapter Three. This technique has been used for the Magnetic Force Microscopy (MFM), and is especially useful for the optical fiber tips used in the Near-field Scanning Optical Microscopy (NSOM).

Cantilever-Based Force Sensors

The cantilever-based force sensor is the sensor used in AFM. Various techniques of detecting microscopic spatial displacement have been developed for the cantilever-based force sensor. Most of these sensing techniques can detect a displacement of the cantilever on the order of one angstrom or less. Since tip-sample interaction forces are usually of the order of a nano-newton, a microscopic cantilever with spring constant of less than 1 newton/meter is a suitable device to convert such microscopic forces into microscopic movement. The processes for large-scale production of microscopic cantilevers by photolithography techniques have been developed.²

Figure 2.2 shows the basic components of a cantilever-based force sensor. It consists of a cantilever, a material stylus, and a displacement sensor. The probe generally has an aspect ratio (which is the ratio of the probe height to the maximum probe width, see Figure 2.2) of greater than one. In most cases, the displacement sensor is external and is located behind the cantilever. When the electromechanical effect (such as piezoelectricity and piezoresistivity) of the material of cantilever is used to detect the cantilever displacement, the cantilever itself can be an internal displacement sensor.

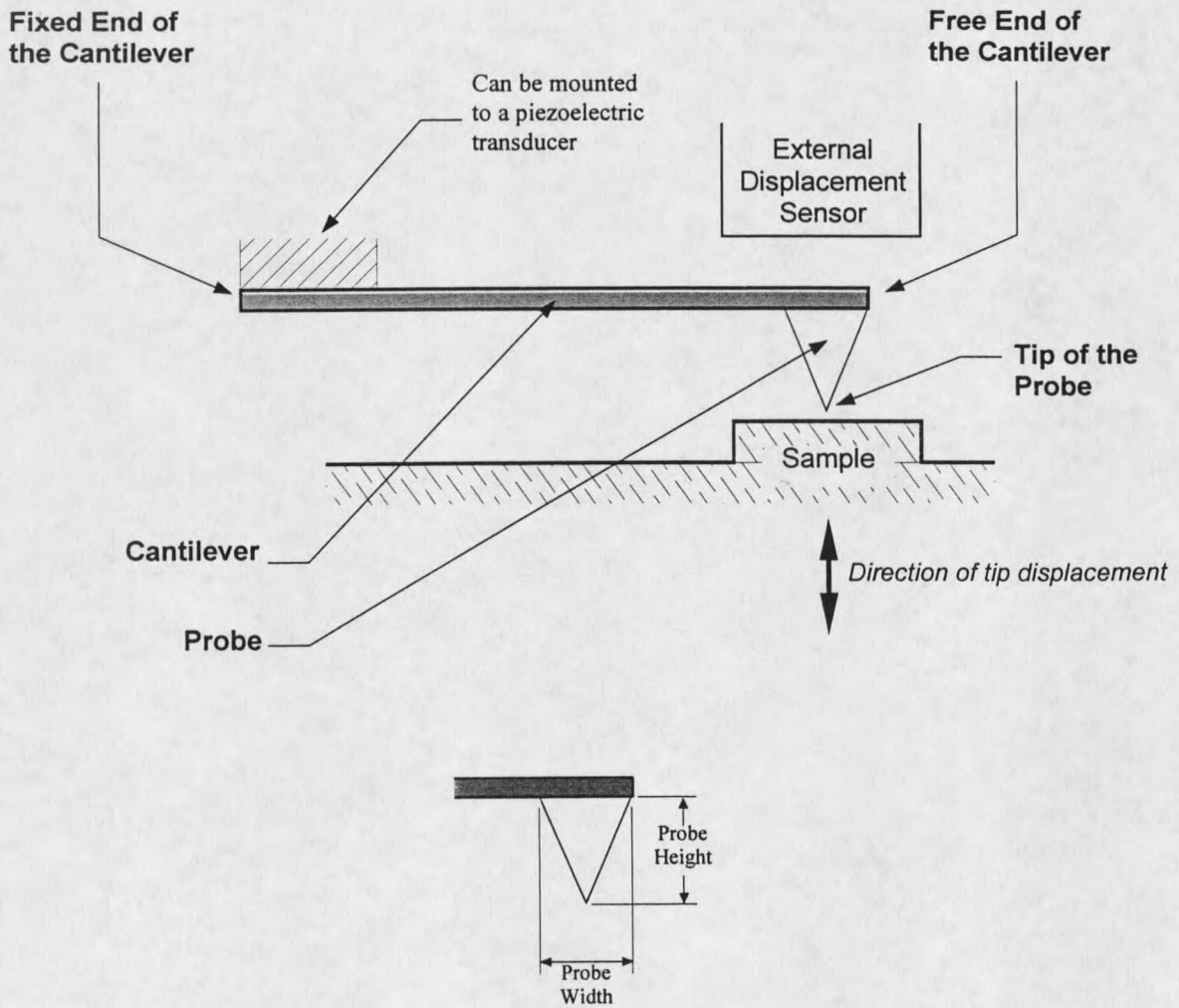


Figure 2.2. Cantilever-based force sensor.

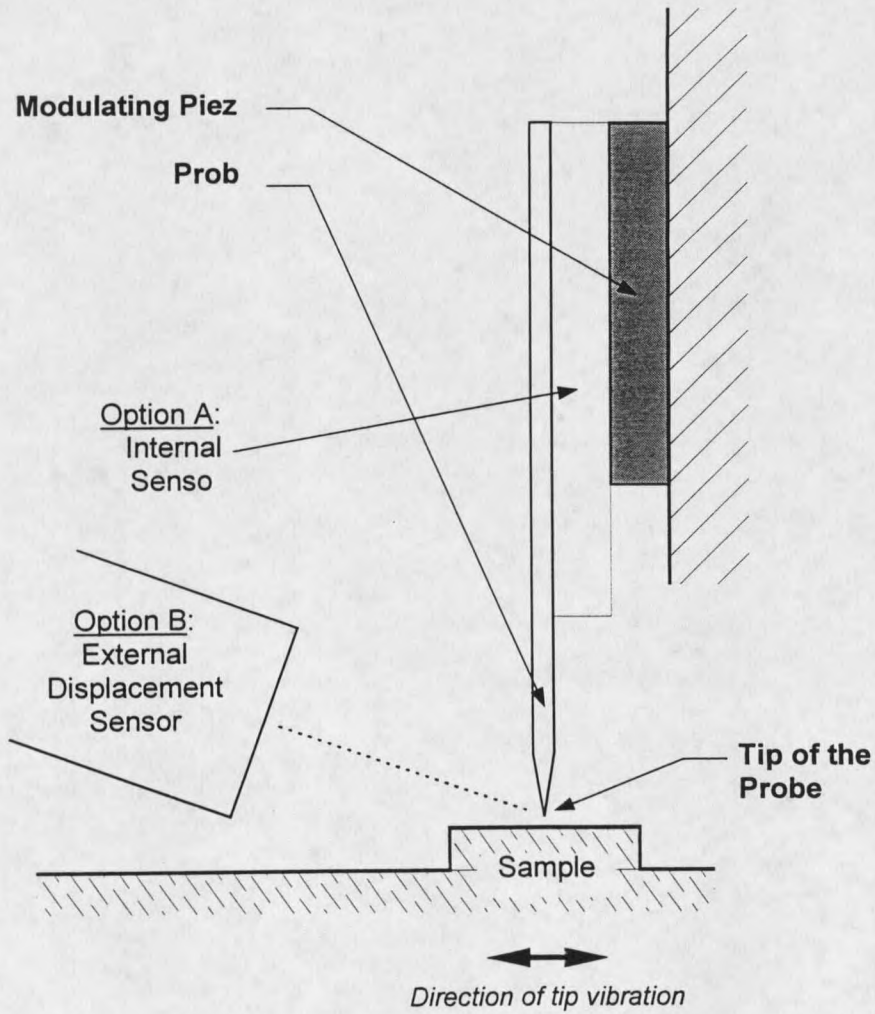


Figure 2.3. Shear force sensor.

Tunneling Sensor. Quantum tunneling was the first phenomenon employed in the design of a nano-newton scale force sensor for the AFM.¹ This force sensor consists of two probes. The first probe that detects the sample is a diamond tip (which is a piece of gold foil 25 μm thick, .25 mm wide, and .8 mm long) mounted on the free end of the cantilever. The second probe is an STM tip, which is used as a displacement sensor to probe the motion of the first probe. It is placed on the other side of the cantilever free end opposite the first probe. To detect the displacement of the cantilever, a tunneling current is established between the STM tip and the metal cantilever. With a fixed bias voltage, the change in tunneling current is a measurement of cantilever displacement.

The advantage of this technique is its high sensitivity, which theoretically can attain 10^{-15} newton at room temperature. However, in order for the tunneling to work, the distance between the cantilever and the STM tip has to be maintained at about 1 nm. This is difficult to achieve when imaging a rough sample. It also has the limitation of not being able to measure lateral forces when the tip is dragged along a scan line.

Capacitance Sensor. If the cantilever is metal or is coated with metal, its nanometer scale displacement can be monitored by the capacitance of the cantilever relative to a fixed reference plate.²⁰

A capacitance sensor has the advantage of easy operation. However, it is difficult to produce such a microscopic capacitor and maintain a small gap. The natural static charge build-up on the cantilever and the reference plate can easily pull the cantilever into contact with the fixed plate, which causes the sensor to stop functioning. Its performance also suffers from the interference of tip-sample capacitance and stray capacitance.

Interferometer Sensor. Using an optical interferometer to detect microscopic displacement is a well-developed technique. This technique has been used to detect the microscopic displacement of an AFM cantilever.²¹ In this kind of sensor, an optical flat is used to form a Fabry Perot interfer-

ometer with the free end of the cantilever. A polarized laser beam is incident on this Fabry Perot. The reflected beam is sent to a photodetector, whose output signal is a sinusoidal function of cantilever displacement. When the cantilever displacement is greater than the wavelength of the laser light, the output signal tracks the displacement into the next fringe. This gives erroneous information about the cantilever displacement, because the same output signal can represent multiple values of cantilever displacement. For this reason, this sensor has difficulty imaging rough samples. Since the distance between the optical flat and cantilever needs to be commensurate with the optical wavelength, the displacement sensor has to be in close proximity to the AFM tip. This makes it difficult to incorporate the AFM with other microscopic techniques, because the displacement sensor will block the line-of-sight of the tip.

Optical Deflection Sensor. When the force between the tip and the sample changes, the cantilever bends to balance the force. In this technique, an optical deflection sensor uses a laser beam to detect the angular change of the cantilever.²² When equipped with a quadrant photosensor, it can detect both normal force and lateral force simultaneously. Because of its high sensitivity and simple instrumentation set up, this is the most widely used technique in AFM today. Because the displacement sensor can be several centimeters away from the tip, it is compatible with various ranging techniques. The major drawback of this sensor is that it requires laser optical alignment whenever a new cantilever is installed in the microscope.

Piezoresist Sensor. Piezoresist sensors are specially engineered cantilevers, which make use of the piezoresist effect of silicon. When the cantilever bends, the internal strain will cause the resistance to change. By measuring the resistance across the cantilever, the interaction force can be detected.²³

The advantage of this sensor is that the cantilever itself is an internal displacement sensor. It allows simple hardware implementation, and no optical alignments are needed. However, because a

finite current must be maintained to measure the resistance, the heating of the cantilever results in the sensor signal being affected by the thermal properties of the sample. When the heat dissipation changes due to the change of surface topography, the piezoresist sensor can create erroneous measurement of the vertical dimension of the surface topography. Because the piezoresist effect is temperature dependent, this force sensor may suffer a large thermal drift during operation.

Shear Force Sensors

Another type of force sensor is the shear force sensor. Instead of utilizing a low-rigidity cantilever like those used in the normal force sensors, it uses a rigid probe with a spring constant on the order of 10^3 newton/meter. A number of shear force sensors have been developed for NSOM, but their capability of controlling tip-sample gap also makes them suitable for AFM.

Figure 2.3 shows the basic components of the shear force sensor. A shear force sensor usually includes a *modulating piezo*, which is a piece of piezoelectric ceramic that supports the force sensing probe and generates vibrations to set the probe assembly into resonance. The probe is perpendicular to the sample surface and the vibration direction is parallel to the sample surface. Since the resonance characteristics of the tip vibration are strongly dependent on the shear force between the tip and the sample, the sample surface can be detected by measuring the change in the resonance characteristics as the tip approaches the sample. With this sensitivity of the sample surface, feedback can be established to measure surface topography.

Compared to the cantilever-based force sensor, non-optical shear force sensors have the advantage of a solid-state structure that allows for easy operation. The rigid probe also ensures a small variation in tip position, providing higher measurement accuracy in surface topography. A drawback of shear force sensors is that it is difficult to track the surface when the slope of the surface topography is too large.

Impedance Sensor. The impedance sensor uses the modulating piezo (see Figure 2.3) itself as

the internal displacement sensor. To detect the tip-sample interaction force, an AC voltage is applied to the modulating piezo to vibrate the probe assembly near its resonance. When the system is in resonance, the AC impedance of the modulating piezo depends strongly on the characteristics of probe assembly's mechanical resonance. Since the resonant characteristics are functions of the tip-sample interaction, measuring the impedance change of the modulating piezo can detect the change in tip-sample interaction.²⁴ The advantage of this sensing mechanism is that it can be simply implemented. However, the piezoelectric ceramics have a strong pyroelectric effect, which generates additional electrical voltages when the temperature changes. For this reason, this technique suffers from thermal stability problems.

Quartz Tuning Fork Sensor. A crystal quartz tuning fork can be used to perform shear force detection.²⁵ In this kind of force sensor, a tuning fork, mounted between the modulating piezo and the probe, is used as the internal force sensor. When the modulating piezo sets the tuning fork into vibration, the piezoelectric effect of the quartz crystal will turn the vibration of the tuning fork into an oscillating electrical signal that can be measured across the electrodes of the tuning fork. The advantage of this technique is its thermal stability and high sensitivity.

Ranging Techniques for Locating Nanometer Scale Objects

Usually, the size of a sample to be studied by a microscope is much larger than the maximum field of view of this microscope. In order to locate the particular area to be studied, two methods can be used. The first is to do a whole area search on the sample with the microscope to look for the area of interest. Obviously, this could be impractical and time-consuming. The second is to use a certain *ranging technique* that incorporates another microscope or imaging instrument that has a greater field of view. In such a greater field of view, the area of interest on the sample surface can be easily located and can be moved into the field of view of the first microscope.

The field of view imaged by a PPM is normally less than 100 μm , which is much smaller than the size of a normal sample. In order for a PPM to be practical, a ranging technique is necessary to locate the sample area to be studied, and then to position the PPM probe over it.

An appropriate ranging technique should have adequate resolution. Its minimum resolvable feature size should be much smaller than the field of view of the microscope. For an optical microscope, the human eye is good enough to be the ranging tool. For the PPM, since its maximum field of view is near the limit of the resolution of a naked eye, some other technique must be used. Optical microscopes have been integrated with a PPM as a ranging tool.²⁶ There are two common methods of integrating an optical microscope: with the optical axis perpendicular to the sample, and with the optical axis at 45° to the sample. With the 90° view, the whole optical field of view is in focus. It is thereby easier to locate an area of interest. With the 90° view, however, the tip of the probe is not visible because the cantilever blocks the line-of-sight. With the 45° view, the tip-sample distance can be monitored easily. This allows fast tip approach for manual control systems but it is not possible to get the entire optical field of view in focus.

There are two shortcomings of a conventional optical microscope that make it unsuitable as a ranging tool for features less than one micrometer. First, the resolution of the conventional optical microscope is limited by the optical wavelength, which makes it impossible to resolve features smaller than about 1 μm . Second, when the magnification is near its maximum, the depth of field of optical microscopes is very small, so it is difficult to locate the area of interest on a very rough sample, such as loose paper fibers.

A desirable ranging tool is the Scanning Electron Microscope (SEM), which usually has a maximum field of view of several millimeters and a resolution of a small fraction of a micrometer. For a field emission SEM, the resolution can be in the nanometer range for certain types of sam-

ples. Integration with a SEM would solve the ranging problems of the PPM. Additionally, the large depth of focus in the SEM permits good focus on both the tip and the sample together at high magnification. A further advantage is that the SEM can be used to monitor the tip condition of a PPM probe.

Since SEM has many capabilities not available from PPM, combining PPM and SEM together in one system will significantly enhance PPM operation. When being monitored by SEM, the PPM probe can be quickly and accurately positioned over a sub-micrometer feature by virtue of visual feedback to the operator. This is important for studying a specific area of the sample and for surface modification at the sub-micrometer scale using the probe. For porous samples with features of a few micrometers or less in size, such as loose fibers, the PPM tip can easily fall into the void space among surface features when not being guided by any ranging tool. In this case, it is necessary to position the tip accurately over a high feature on the sample in order for the relatively short PPM probe to reach a surface for acquiring a PPM image.

Combining STM/AFM and SEM

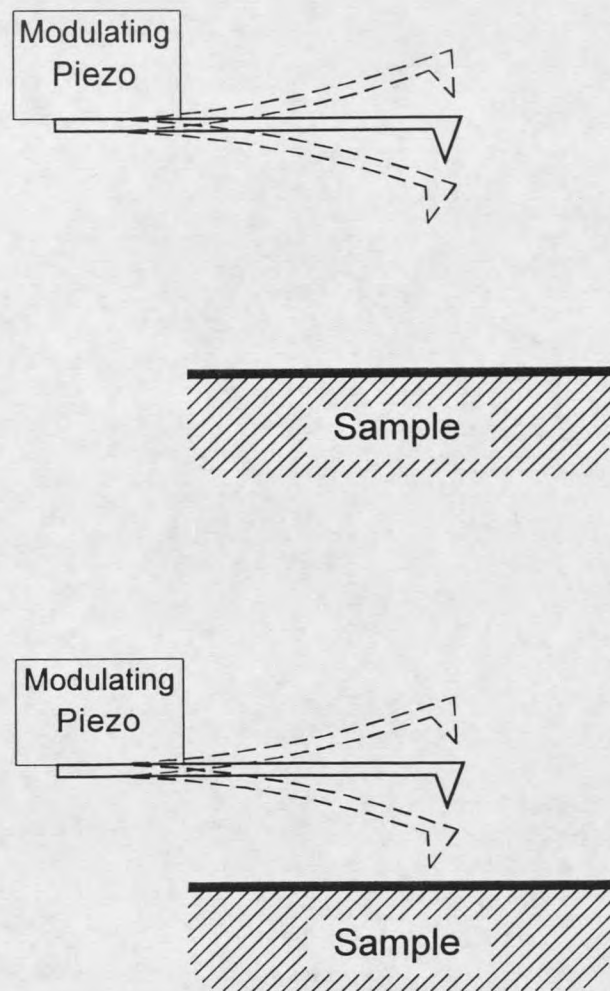
Many efforts have been made to combine the Scanning Tunneling Microscope (STM) with the Scanning Electron Microscope (SEM).^{27,28} A number of these systems were ultra-high vacuum (UHV) STM/SEM systems that operate in a vacuum of about 10^{-10} torr. In the UHV environment, the problem of electron-beam-induced surface contamination of a cleaned sample is minor. However, the shortcoming of these UHV STM/SEM systems is that they are difficult to operate and can only image conducting samples.

Besides the advantage of imaging non-conducting samples, the AFM can perform various types of surface proximity measurements. Some examples of these special working modes are: lateral force microscopy (or frictional force microscopy), magnetic force microscopy (MFM), electri-

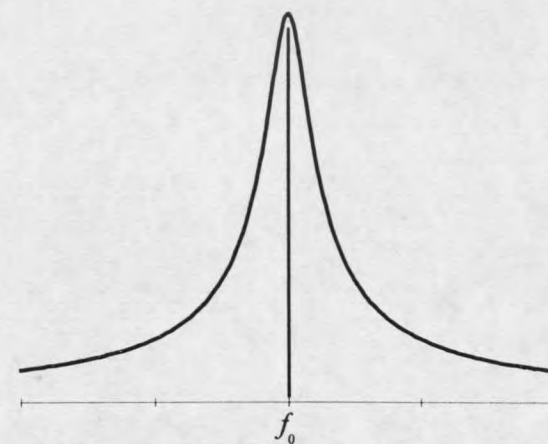
cal force microscopy (EFM), thermal force microscopy, and so on. Because of the advantages of AFM, it is also important to develop an AFM that is equipped with a SEM as a ranging tool. There have been instances of integration of SEMs with AFMs, but not for the purpose of ranging. An AFM/SEM system has been built as a station for electron-beam tip fabrication and *in situ* tip testing.²⁹ In this system, the sample faces downward. When performing AFM imaging, the AFM tip and the AFM imaging area on the sample cannot be imaged with the SEM, preventing the SEM from being used as a ranging tool. Another AFM/SEM system has used electron-beam deflection to sense cantilever bending.³⁰ A piezo lever AFM has been operated simultaneously with a SEM,³¹ which demonstrated the compatibility of the piezoresist lever with SEM. However, it did not have any translators, nor could it be used as a ranging tool. Our combined AFM/SEM system was the first full-feature instrument that allowed simultaneous AFM and SEM operations to monitor the scanning of the AFM tip, and to accurately position the tip over a sub-micrometer scale feature.³²

Vibrating Cantilever Mode of AFM Operation

An important AFM operating mode is the Vibrating Cantilever Mode (VCM). The concept of using a vibrating probe to measure surface profile was first introduced in the profiler instruments over 40 years ago.³³ In vibrating cantilever AFM, the fixed end of the cantilever is driven with a modulating piezo, so that the cantilever-tip assembly is vibrated continuously perpendicular to the sample surface. Figure 2.4 shows the working principle of VCM. When the tip is far away from the sample (Figure 2.4a), the cantilever has a defined resonant frequency f_0 . When the tip gets close to the sample (Figure 2.4b), the tip-sample interaction causes changes in resonant characteristics of the cantilever (see Chapter Five). Particularly, the resonant frequency changes to f'_0 . If the driving frequency of the cantilever is set at f_0 , the electronic system will detect a change in the vibration amplitude, ΔA , when the tip is interacting with the sample. A similar effect also happens to the phase shift signal. Therefore, the sample surface can be detected by the changes in vibration amplitude and phase shift.



(a)



(b)

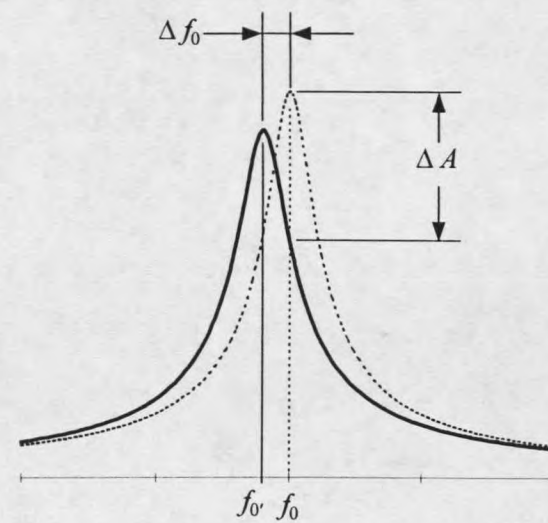


Figure 2.4. The working principle of the AFM vibrating cantilever mode (VCM).

If the probe is not in vibration during the scanning process, the tip would be continuously in contact with the sample, and the lateral force (friction) between the tip and the sample would damage the tip and/or the sample surface. The primary reason for oscillating the probe perpendicular to the sample surface while scanning is to reduce the unwanted lateral forces between the probe tip and sample surface; thus minimizing the tip and/or sample damage. To achieve these benefits, the probe in a profiler instrument is typically oscillated between the point of contact and a position well above the sample's surface.³³ However, since the contact force in profiler instruments is relatively large, the lateral resolution (see Chapter Three) of surface topography is on the order of one micrometer.

A dramatic improvement in the lateral resolution and force sensitivity of profiler instruments occurred with the development of AFM. As early as 1986, Binnig and Quate acknowledged the potential benefits of vibrating the cantilever in an AFM.¹ Martin et al.⁶ and later Meyer²² demonstrated the importance of vibrating techniques applied to AFM with a feedback mechanism based on monitoring the changes in amplitude or phase of a vibrating cantilever. Martin et al.⁶ demonstrated two modes of operation, a low-amplitude mode and a high-amplitude mode.

The low-amplitude mode operates with low vibration amplitudes, which allow the probe to profile the surface with high resolution without surface contact. The cantilever vibrates at or near its resonant frequency at relatively low vibration amplitudes, on the order of 2 nm or less. It is believed that the tip is working in the van der Waals force regime of the tip-sample interaction potential curve. In this regime, the force is attractive and the force gradient is positive (the attractive force increases as the tip approaches the sample). Such a tip-sample interaction effectively reduces the spring constant of the cantilever, and lowers the resonant frequency. With a fixed driving frequency, the changes in resonant frequency result in the changes of the vibration amplitude or the phase shift (see Chapter Three), which can be detected and used to set up a feedback maintaining a constant average tip-sample gap. The feedback establishment can also be done by detecting the change of resonant frequency.³⁴ This scanning mode has come to be known as the non-contact

mode AFM. Ideally, with non-contact imaging, no damage should occur to the sample surface and the AFM tip, that is, the imaging process is non-destructive.

In ultra-high vacuum conditions, where samples can maintain less than one monolayer of surface contamination for a few hours, atomic resolution has been achieved on Si(7x7)^{35, 36} and InP(110)³⁷ surfaces. The stable atomic resolution images indicate that the imaging process is non-destructive to both the tip and the sample. Although the non-contact mode has been very successful in UHV, it has not been demonstrated capable of providing high resolution images in ambient air. A widely accepted argument links the poor resolution with the surface contamination layer. When used under ambient conditions, the surface contamination layer between the tip and the sample results in large tip-sample gap and creates instability in the feedback loop. Both of these factors lower the lateral resolution of AFM (see Chapter Five). Also, it is believed that the capillary force of this viscous layer on the surface can "capture" the tip to the surface, so that non-contact imaging is not possible.⁸

Martin et al.⁶ have demonstrated a second vibrating mode using an additional relatively large oscillation, approximately 100 nm in amplitude. This high-amplitude tip oscillation is at a relatively low frequency, which is added to the low-amplitude vibration at the cantilever resonant frequency. In this high-amplitude oscillation AFM mode, the tip contacts the surface upon each low frequency oscillation. Because the tip contacts the surface, damage to the tip and/or sample surface is possible. On the other hand, some researchers believe that little sample and tip damage occurs when the probe tip contacts the sample in the AFM vibrating cantilever modes.⁸ This has not been proved experimentally.

Nanometer Scale Surface Electrical Potential Sensors

When imaging with a conductive tip in AFM vibrating cantilever mode, variations in surface potential or localized static charges can be imaged along with the topography. Microscopic imag-

ing of potential distribution is important for analyzing characteristics of semiconductor devices. Microscopy of surface potential distribution can also be applied to organic and biological systems.

Kelvin-probe Force Microscopy

The first PPM method developed for direct imaging of surface potential is based on the traditional Kelvin method. The Kelvin method is a simple technique for measuring contact potential difference between two surfaces.³⁸ In the Kelvin method, the probe and the sample form a capacitor. When the probe vibrates the capacitance changes, and since the contact potential is constant, a current is set up in the circuit according to the equation,

$$i(t) = \frac{dQ}{dt} = \frac{d}{dt}(CV) = V \frac{dC}{dt}. \quad (2.1)$$

Since the AC current is the result of the periodic change of the capacitance, the Kelvin method is also called the vibrating capacitor method.

To perform a surface potential measurement with high lateral resolution, the Kelvin method was incorporated with an AFM,³⁹ based on the vibrating cantilever mode, which is called *Kelvin-probe Force Microscope* (KFM). In this method, instead of driving the cantilever to vibrate mechanically by a modulating piezo, it is driven electrically by an AC voltage applied to the conducting probe. With this external AC bias voltage, any potential difference between the probe and sample will cause the cantilever to vibrate. (The details of the working mechanism will be shown in the following paragraphs.) The reason for this design change to the original Kelvin method is that measuring mechanical movement of a cantilever is what an AFM force sensor does best. In the traditional Kelvin method, probe vibration is used to generate an oscillating current, which is then measured. In the Kelvin-probe force microscopy, an AC bias voltage is used to generate a probe vibration, which is then measured.

To show the details of the KFM working mechanism, we first consider the traditional Kelvin method, which is commonly used to measure contact potential between two materials. The contact

potential difference between the two materials is $V_{CPD} = 1/e(\phi_2 - \phi_1)$, where ϕ_1 and ϕ_2 are the work functions (in the unit of eV) of the two materials. When the distance between the two materials is oscillating at the frequency ω , the change in capacitance, ΔC , will generate a current of³⁹

$$i(t) = V_{CPD} \omega \Delta C \cos \omega t. \quad (2.1)$$

The contact potential difference is measured by applying a sinusoidal vibration between the two materials to change the capacitance periodically. The change in capacitance generates an AC current, $i(t)$. Finally, a bucking voltage is applied to make $i(t) = 0$. Then the externally applied voltage is equal to the V_{CPD} .

In applying the Kelvin method to AFM, when the cantilever-probe assembly gets close to the surface, the contact potential difference between the tip and the sample results in an electrostatic force which can then be detected by the AFM cantilever. The electrostatic force between two conductive surfaces can be written as⁴⁰

$$F = -\left(\frac{1}{2}\right) V^2 \left(\frac{\partial C}{\partial G}\right), \quad (2.2)$$

where C is the capacitance between the tip and the sample, G is the tip-sample gap, and V is the potential difference.

To measure the surface electrical potential, both AC and DC voltages are applied to the conducting AFM probe from power supplies. The total voltage on the tip is,

$$V = V_{DC} + V_{AC} \sin(\omega t), \quad (2.3)$$

where V_{DC} includes the surface potential difference to be measured, and $V_{AC} \sin(\omega t)$ is the AC voltage. In the measurement, the amplitude of cantilever movement is small relative to the size of the cantilever. Therefore, we can assume the vibration amplitude is proportional to the force. From equation (2.2) and (2.3), the vibration amplitude of the cantilever, A , can be written as⁴⁰

$$A = S \left[V_{DC}^2 + 2 V_{DC} V_{AC} \sin(\omega t) + \frac{V_{AC}^2}{2} (1 - \cos(2\omega t)) \right], \quad (2.4)$$

where S is a coefficient that includes the effect of $\partial C/\partial G$, surface chemistry, and scan conditions.

The potential imaging is performed by measuring the $\sin(\omega t)$ component, which is the second term in equation (2.4). An external DC voltage is applied to cancel the contact potential difference, making $V_{DC} = 0$. Then the second term in the square brackets vanishes and the vibration at the fundamental frequency will thus vanish. At this point, the surface potential difference is equal to the external DC voltage on the tip.

KFM instrumentation consists of two basic parts incorporating two different feedback loops. The first part is a regular vibrating cantilever AFM, which makes up the topographic feedback loop that keeps the tip-sample gap constant. It consists of a cantilever mechanically vibrating at frequency ω_1 , which is the working frequency of the topographic feedback loop. The second part is the Kelvin probe, which makes up the electrical potential feedback loop that keeps the DC voltage between the tip and the sample constant. It supplies an electrical AC voltage of frequency ω_2 , which is the working frequency of the potential feedback loop, to the metal (or metalized) cantilever. The operation frequency for the topographic feedback is usually set at the resonant frequency of the cantilever, and the operation frequency for the potential feedback is about 2 kHz above the resonance.³⁹

Maxwell Stress Microscopy

Another technique for imaging surface potential, developed shortly after the invention of KFM, is Maxwell Stress Microscopy, which is a technique modified from KFM. The difference is that the frequency used for the topographic feedback loop is the second harmonic, 2ω , of the resonance.⁴¹ From the equation (2.4), we see that when $V_{DC} = 0$, only the second harmonic term, 2ω , is left. This term is independent of the DC bias between the tip and the sample. Therefore, when topographic feedback is working on the second harmonic, the feedback condition can be totally independent of surface potential.

In KFM, the working frequencies of the two feedback loops are both near the first harmonic, which could cause interference between the two feedback loops. The advantage of Maxwell Stress

Microscopy is that it can completely remove the crosstalk between the topographic signal and the potential signal. The drawback is that its topographic sensitivity is very low, because the vibration amplitude of the second harmonic is significantly smaller than the amplitude of the first harmonic. In this case, the signal-to-noise ratio is small, which reduces the sensitivity of the topographic feedback loop. Therefore, this method is difficult to use on samples with rough structures, such as a step feature on an integrated circuit specimen.

CHAPTER 3

THEORETICAL ANALYSIS

Introduction

In the following discussion, we first consider the interactions between two neutral objects, where we will compare the magnitude of the van der Waals force with the capillary force. Then, we discuss the lateral resolution with an example of imaging a single charge. To study the operation of a vibrating cantilever, we present the basic information of mechanical resonance with viscous damping. The largest section in this chapter is devoted to the analysis of tip-sample contact, which is an important factor in AFM operations. At the end we present the expression of the surface potential on a semiconductor junction, which will be applied in a KFM measurement in Chapter 7.

Nanometer Scale Surface Interactions

Interactions in the nanometer scale can be classified into a number of types depending on the actual systems involved. Although various force names are used for these interactions, they are all manifestations of the electromagnetic force. The most well known interaction is the van der Waals force under ideal conditions. Under ambient air conditions, since the surface is covered with a layer of liquid-like contamination, the capillary force is dominant when a liquid bridge is formed between the two interacting surfaces. For normal PPM operations, the distance between the tip and the sample is usually less than 10 nm. Studying various nanometer scale tip-sample interactions is critical to understanding the imaging mechanisms of PPM.

Interaction between Neutral Molecules

The attractive interaction between neutral molecules can be described with the van der Waals force. The major characteristic of the van der Waals force is that its potential energy depends on the inverse sixth power of the distance. The van der Waals force can be contributed by any one of the following three components.⁴² The first is the dipole-dipole interaction that is referred to as the *orientation* or *Keesom* interaction. The second is the dipole-induced dipole interaction that is referred to as the *induction* or *Debye* interaction. The third is the dispersion forces that are the result of interaction among the instantaneous dipoles of all the atoms in the interacting objects. All of these interactions follow the inverse sixth power of distance dependence, that is,

$$w(r) \propto 1/r^6. \quad (3.1)$$

In general, the interaction between two neutral molecules involves both attractive and repulsive forces. The general form of this potential is called the Mie potential,⁴³

$$w(r) = -\frac{A}{r^n} + \frac{B}{r^m}, \quad (3.2)$$

where A and B are constants that depend on the specific system. The most important repulsive force between the ion cores of the two atoms⁴⁴ is due to the Pauli's exclusion principle when the gap between them is on the order of 0.1 nm or less. Since the repulsive force is greater than the attractive force at short distance, we have $m > n$. The commonly used form is the *Lennard-Jones potential* (or *6-12 potential*⁴⁴) which combines the inter-atomic repulsion and the van der Waals attraction, that is⁴⁵

$$\begin{aligned} w(r) &= -\frac{A}{r^6} + \frac{B}{r^{12}} \\ &= 4w_0 \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right], \end{aligned} \quad (3.3)$$

where w_0 is the minimum value of the potential energy. The force is

$$F(r) = -\frac{dw(r)}{dr} = 24w_0 \left[2 \left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^6 \right] \frac{1}{r}. \quad (3.4)$$

The Lennard-Jones potential is commonly used to describe the interaction between two neutral particles. The first and second order derivatives are

$$\frac{dF(r)}{dr} = 24w_0 \left[-26 \left(\frac{\sigma}{r} \right)^{12} + 7 \left(\frac{\sigma}{r} \right)^6 \right] \frac{1}{r^2}, \quad (3.5)$$

and

$$\frac{d^2 F(r)}{dr^2} = 672w_0 \left[13 \left(\frac{\sigma}{r} \right)^{12} - 2 \left(\frac{\sigma}{r} \right)^6 \right] \frac{1}{r^3}. \quad (3.6)$$

From equation (3.4) with the condition of $F(r_0) = 0$, we have the inter-molecular separation where the potential energy is minimum, that is $w(r_0) = -w_0$,

$$r_0 = \sqrt[6]{2}\sigma = 1.122\sigma. \quad (3.7)$$

From equation (3.5), with the condition $dF/dr(r = r_a) = 0$, we get the inter-molecular separation where the magnitude of attractive force is maximum,

$$r_a = \sqrt[6]{\frac{2 \times 13}{7}}\sigma = 1.244\sigma. \quad (3.8)$$

Combining equations (3.4) and (3.8), we have the maximum attractive force,

$$F(r_a) = -\frac{96}{13} \left(\frac{7}{26} \right)^{\frac{7}{6}} \frac{w_0}{\sigma} = -1.598 \frac{w_0}{\sigma}. \quad (3.9)$$

From equation (3.6), with the condition $d^2 F/dr^2(r = r_s) = 0$, we get the inter-molecular separation where the force gradient is maximum,

$$r_s = \sqrt[6]{\frac{13}{2}}\sigma = 1.366\sigma. \quad (3.10)$$

Combining equations (3.5) and (3.10), we have the maximum force gradient,

$$\frac{dF}{dr}(r_s) = 72 \left(\frac{2}{13} \right)^{\frac{4}{3}} \frac{w_0}{\sigma^2} = 5.94 \frac{w_0}{\sigma^2}. \quad (3.11)$$

Figure 3.1 shows the force presented by equation (3.4), and the three important values of the inter-atomic separation, r_0 , r_a and r_s .

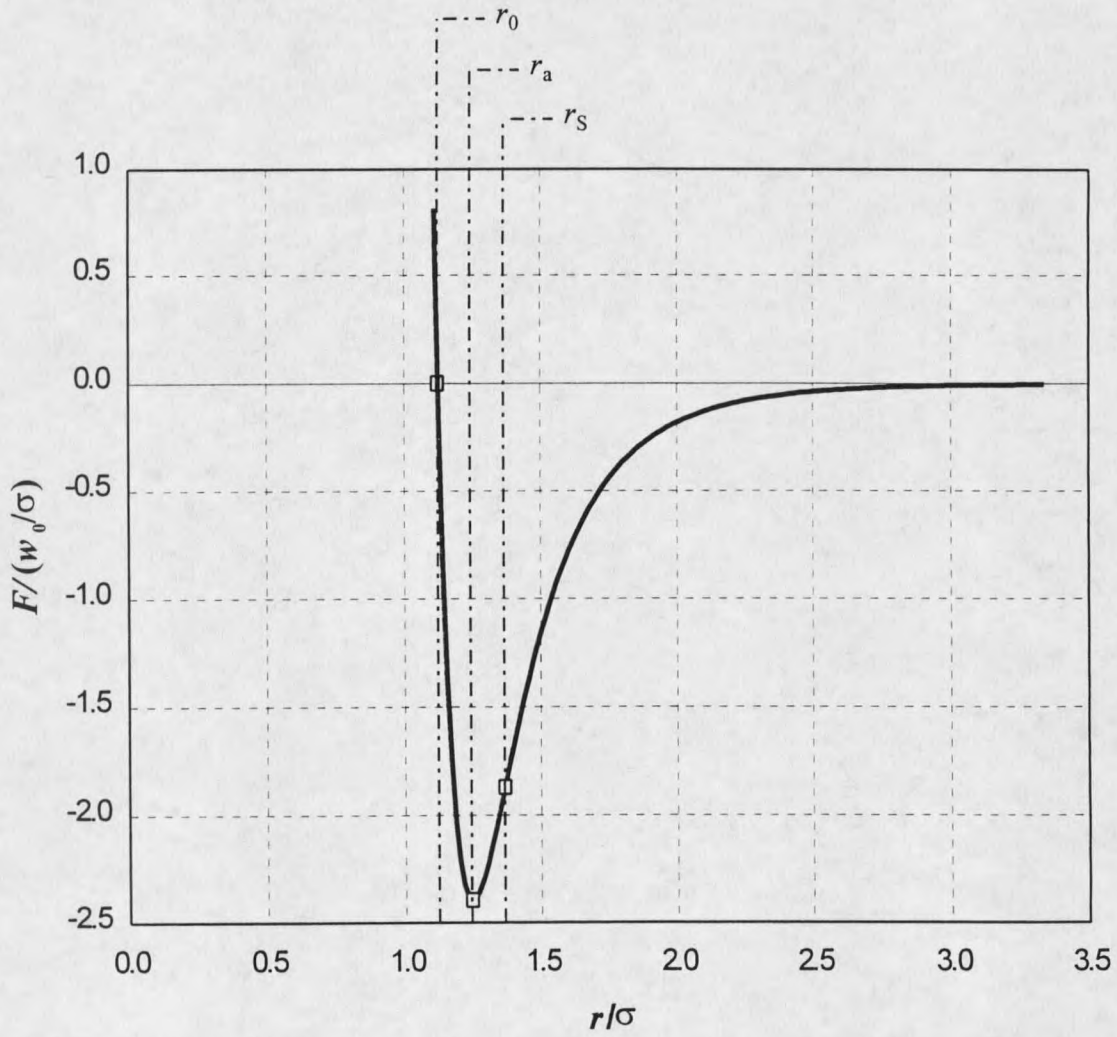


Figure 3.1. The force created by the Lennard-Jones potential. r_0 is the spacing where the net force is zero. r_a is the spacing where the maximum attractive force occurs. r_s is the spacing where the force gradient is maximized.

Interaction between a Sphere and a Flat Surface

The 6-12 potential described above is only for the interaction between two neutral particles with negligible volume. In the reality of tip-sample interaction in PPM, the physical dimensions are significant for the tip-sample interaction. Furthermore, for PPM operation in air, due to surface contamination (mostly the condensation of water) on both the tip and the sample surfaces, the capillary force between the tip and the sample should also be considered. If static charges exist on the surfaces, electrostatic interaction occurs, which usually dominates at long range. When the tip makes physical contact with the sample, the two materials can be deformed by the high pressure in the contact region, which results in the repulsive contact force. Therefore, to study the tip-sample interaction in air, we should consider four types of interactions: van der Waals force, capillary force, electrostatic force and repulsive force due to pressure deformation of the surface. Since our studies are focused on the neutral tip-sample interaction prior to physical contact, only the van der Waals force and the capillary force are discussed in the following sections.

van der Waals Force. To study the interaction between the tip and the sample, we will use a simplified model that the tip is a sphere of radius R and the sample is a flat surface. The van der Waals potential between a sphere and a flat surface is⁴⁶

$$w(G) = -\frac{HR}{6G}, \quad (3.12)$$

where G is the tip-sample gap and H is the conventional Hamaker constant given by⁴⁷

$$H = \pi^2 A \rho_1 \rho_2, \quad (3.13)$$

where A is the coefficient in equation (3.3) and ρ_1 and ρ_2 are the number of atoms per unit volume in the two interaction bodies. From equation (3.12), we have the attractive force,

$$F_{vdW} = -\frac{HR}{6G^2}. \quad (3.14)$$

Capillary Force. When the tip and the sample make contact in air, a meniscus forms around

the contact region (Figure 3.2). With the assumption that the contact area is much smaller than the size of the tip, the capillary attraction force due to the liquid bridge is⁴⁸

$$F_{Cpl} = -\frac{4\pi R\gamma_L \cos(\theta)}{1 + \frac{G}{d}}, \quad (3.15)$$

where R is the tip radius, γ_L is the surface energy of the liquid, θ is the contact angle, and d is the dimension illustrated in Figure 3.2. The volume of the liquid between the two objects is

$$V \cong \pi(R \sin \phi)^2 (G + d) - \pi R d^2 + \frac{\pi}{3} d^3, \quad (3.16)$$

where ϕ is the angle shown in Figure 3.2. Since d is very small, we can ignore the last two terms in the above equation, and have

$$G + d = G_0 = \text{constant}. \quad (3.17)$$

Combining equations (3.15) and (3.17), we have

$$F_{Cpl} \cong -F_0 \left(1 - \frac{G}{G_0}\right), \quad (3.18)$$

where $F_0 = 4\pi R\gamma_L \cos(\theta)$ is the maximum capillary attractive force, and G_0 is where the capillary force vanishes.

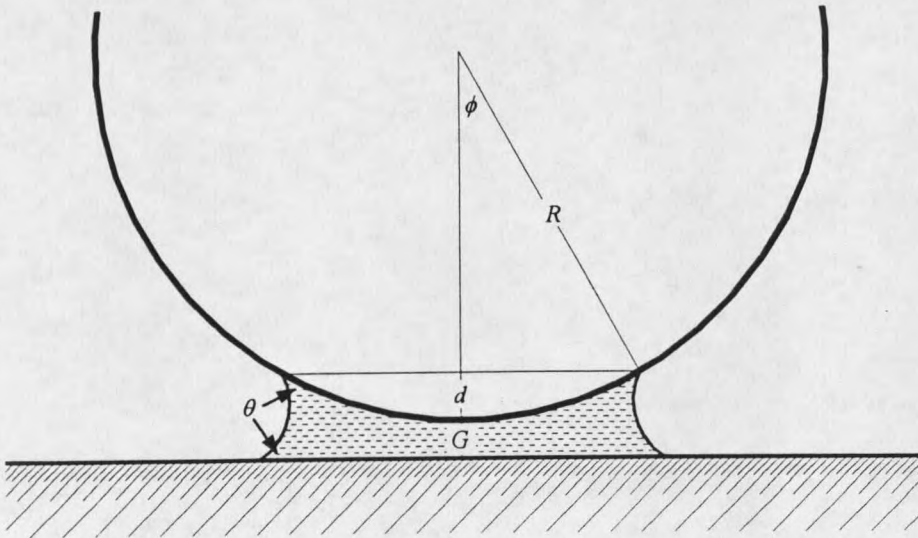


Figure 3.2. The capillary attraction between a sphere and a flat surface.

Comparing van der Waals Force with Capillary Force. To get a more physical understanding of the van der Waals force and the capillary force, we will make some estimation based on some realistic numbers. Since the Hamaker constants of most condensed phases are found to lie in the range $(0.4-4)10^{-19}$ J,⁴⁹ we will assume a value of $H = 1 \times 10^{-19}$ J when calculating the van der Waals force.

To calculate the capillary force, we use $\gamma_L = 72$ mJm⁻² for water,⁵⁰ choose $\cos(\theta) = 0.5$, and assume the total thickness of the contamination layers on both surfaces to be $G_0 = 10$ nm, which is a common value observed in our studies. From equations (3.14) and (3.18), the forces are calculated with two different values of sphere radius and shown in Figure 3.3.

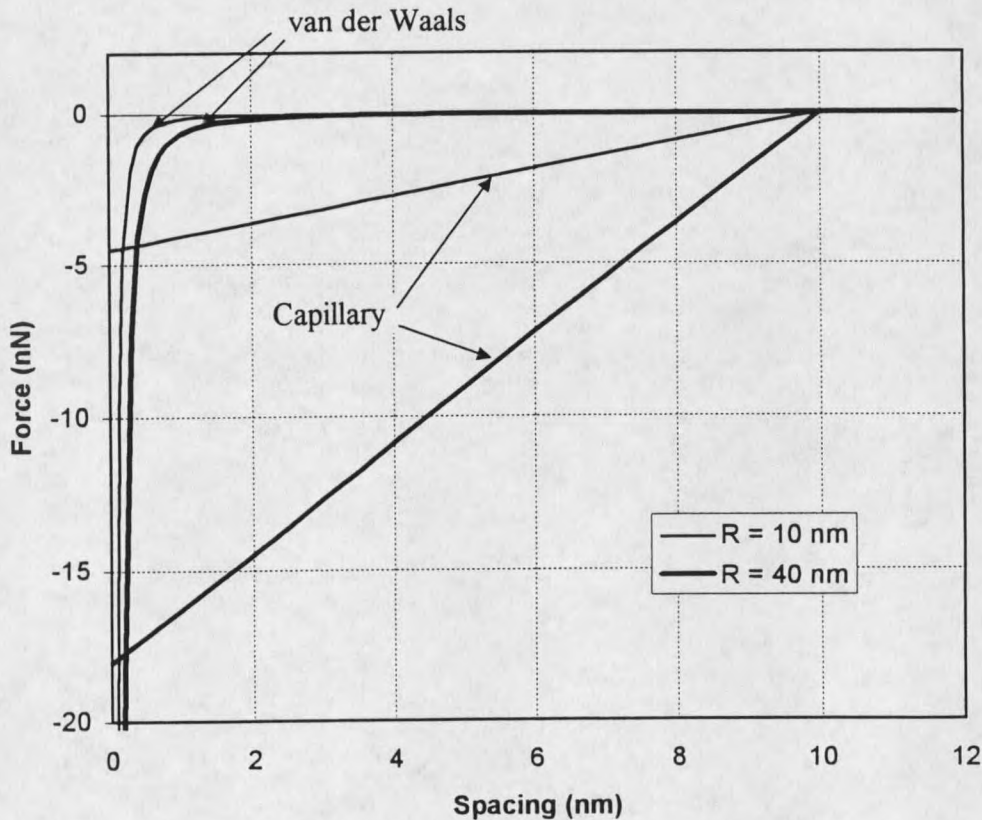


Figure 3.3. Comparison of the van der Waals force and capillary force between a sphere and a flat surface for two different values of the radius, R .

Lateral Resolution in Imaging the Electrostatic Properties

In microscopy, *lateral resolution* generally means the smallest feature that can be resolved by a microscope. In an optical microscope or a SEM, it is determined by the smallest focus spot. In PPM, the lateral resolution of an image is governed by the tip radius and tip-sample gap. Particularly, in STM, the lateral resolution has been defined by a range around the tip where the change in the tunneling current is less than one order of magnitude.⁵¹ In force microscopy, the lateral resolution can be defined by the change in tip-sample interaction forces. In this section, we will analyze the electrostatic interaction based on a simple model. The lateral resolution will be defined, and its relationship with tip parameters will be shown.

When measuring surface electrostatic properties, a conducting tip is always used. Since the lateral resolution is a one-dimensional parameter, a two-dimensional model is sufficient for the study. Figure 3.4 shows a theoretical model for the following analysis. We assume that the tip is a spherical grounded conductor with radius R , and the tip-sample gap is G . On the sample surface, there is a single point charge q that is displaced laterally x from the tip. This model pertains to a situation in which the electrostatic force is the major tip-sample interaction.

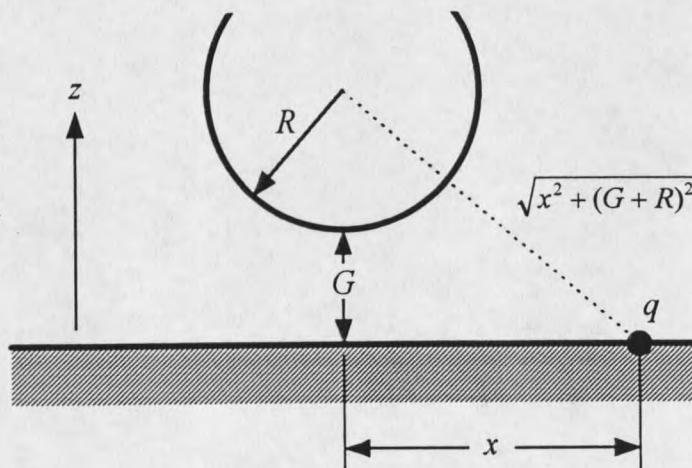


Figure 3.4. Diagram of a single charge and a conducting sphere.

Since the vibrating cantilever AFM detects mostly the force component perpendicular to the sample surface, we will study the vertical component of the force between this physical charge on the surface and the image charge induced in the tip. Generally, this force component is a function of x , that is $f = f(x)$. When the charge is right under the tip, the force is at its maximum. That is, $df/dx(x=0) = 0$, and $f_{MAX} = f(0)$. To define the lateral resolution, let us consider two point charges that are at location x_1 and x_2 respectively. The total force applied to the tip when the tip is scanning along the x direction is

$$f_T(x) = f(x-x_1) + f(x-x_2). \quad (3.19)$$

To define the lateral resolution is to define a minimum resolvable size, Δ_{Lat} , which is illustrated in Figure 3.5. Any feature smaller than Δ_{Lat} is considered unresolvable. The minimum resolvable size, Δ_{Lat} is defined by the relation,

$$f\left(\frac{\Delta_{Lat}}{2}\right) = \frac{1}{2} f(0). \quad (3.20)$$

In the case illustrated in Figure 3.5, when the distance between the two charges is smaller than Δ_{Lat} , they are considered not resolvable as a two-charge feature. When the minimum resolvable size is small, the lateral resolution is high.

Next, let us find the specific expression of this force. The attractive electrostatic force between a point charge and the conducting sphere is⁵²

$$|\bar{F}(x, z, R)| = q^2 \frac{R\sqrt{x^2 + (z+R)^2}}{(x^2 + z^2 + 2zR)^2}, \quad (3.21)$$

and its vertical component is

$$F_z(x, z, R) = q^2 \frac{R(z+R)}{(x^2 + z^2 + 2zR)^2}. \quad (3.22)$$

This is the force detected by the cantilever, that is, $f(x) = F_z(x, z, R)$.

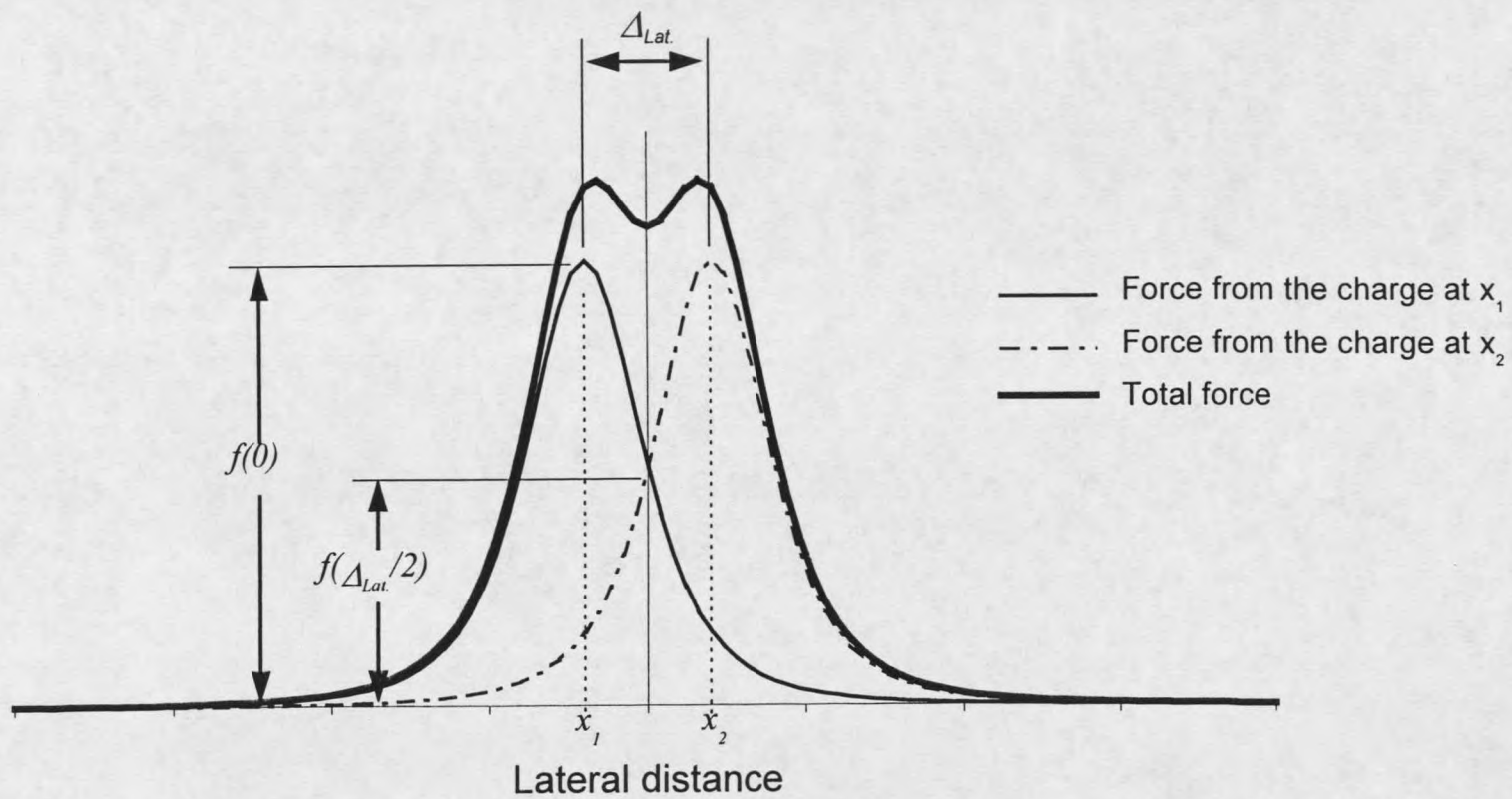


Figure 3.5. The definition of lateral resolution.

Obviously, the force that the cantilever detects is also a function of tip-sample gap, G , and the tip radius, R . This dependence is shown in Figure 3.6. From these diagrams, we can get some information. When the tip is very close to the sample, the maximum force (where $x = 0$) is not dependent on the radius of the tip. That is,

$$F_z(0, G) = q^2 \frac{R(G+R)}{G^2(G+2R)^2} \xrightarrow{G \ll R} \frac{q^2}{4G^2}. \quad (3.23)$$

This means that the strongest interaction applied to the tip is mostly on the very bottom part of the tip.

We can also draw some conclusions on the minimum resolvable size. From equation (3.20), we have the minimum resolvable size,

$$\Delta_{Lat.} = 2 \sqrt{\left(\sqrt{\frac{1}{0.5}} - 1 \right) G(G+2R)}. \quad (3.24)$$

This equation shows clearly the dependence on the tip-sample gap, G , and the tip radius, R . From the above equation, the minimum resolvable size can also be expressed as

$$\Delta_{Lat.} \propto \sqrt{R}, \quad (3.25)$$

and

$$\Delta_{Lat.} \propto G. \quad (3.26)$$

These equations indicate that reducing both R and G will reduce $\Delta_{Lat.}$. However, the minimum resolvable size, $\Delta_{Lat.}$, has a stronger dependence on the tip-sample gap, G , than the tip radius, R . In order to get higher lateral resolution, it is more important to reduce the tip-sample gap than to reduce the tip radius.

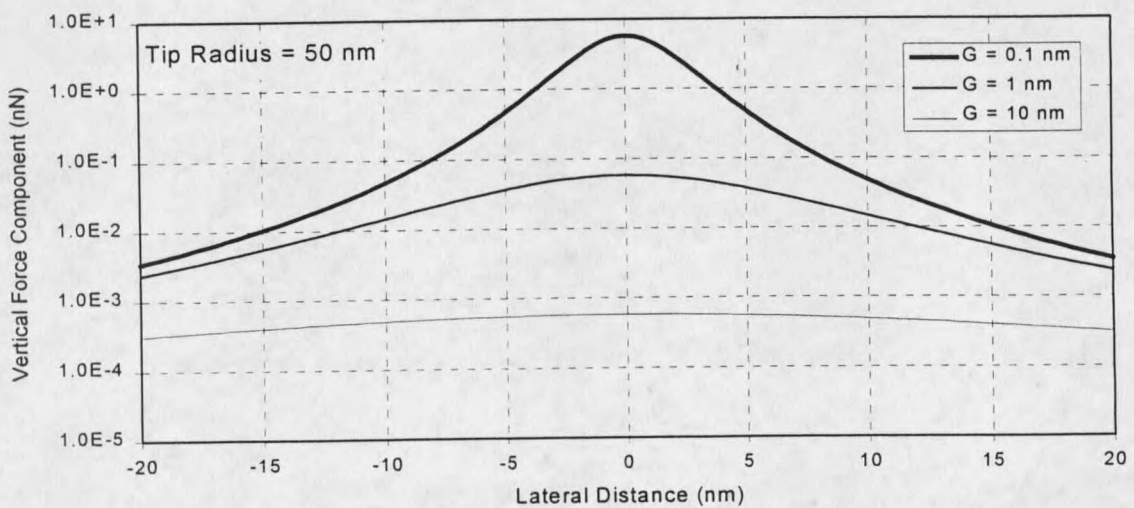
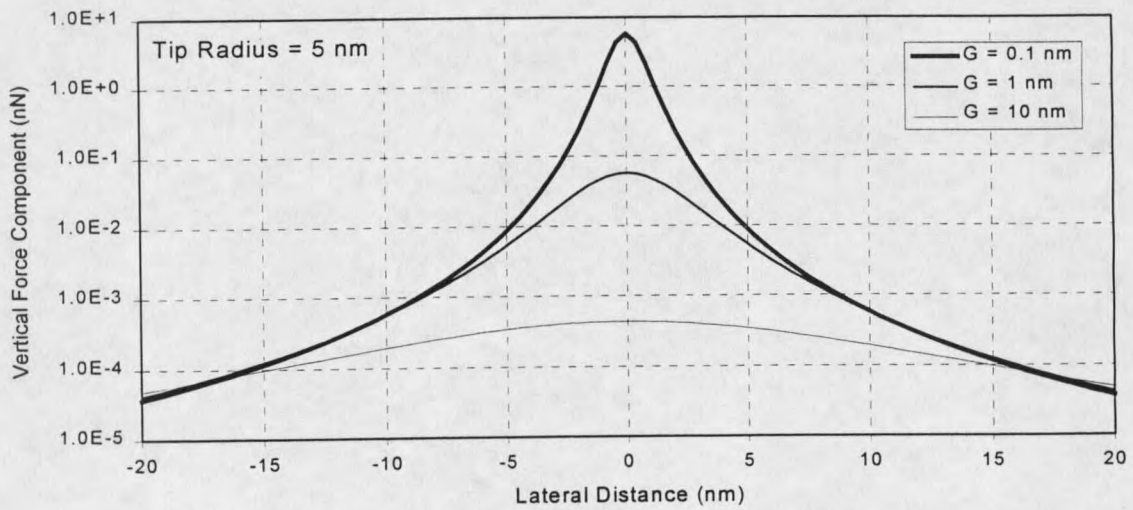
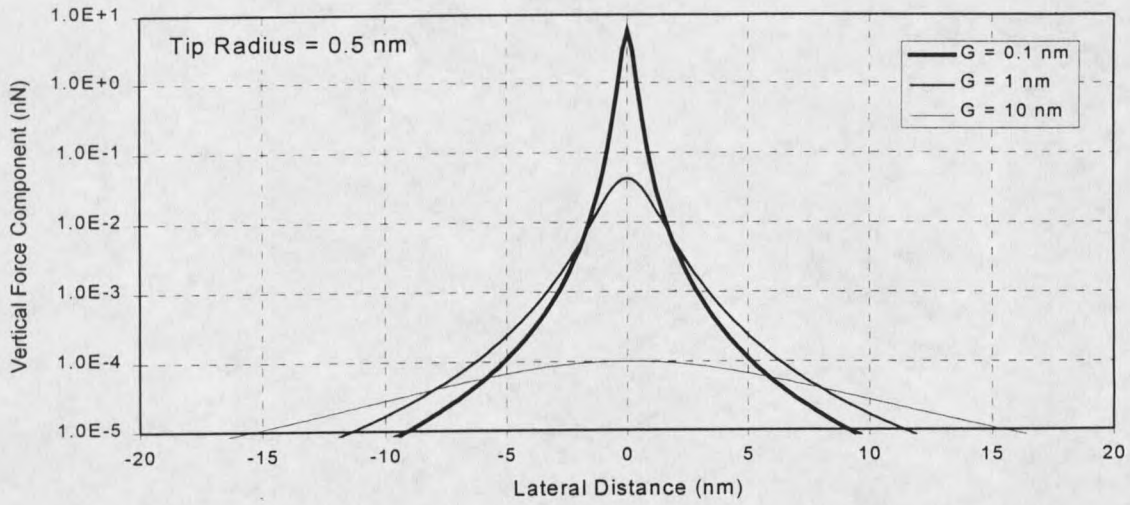


Figure 3.6. Electrostatic force between a single charge and a conducting tip.

Mechanical Resonance of a Harmonic Oscillator with Viscous Damping

In vibrating cantilever mode AFM operation, the damping of the cantilever vibration is due to the collision with air molecules and the dissipation from the cantilever deformation. In normal AFM operation, the amplitude of the cantilever vibration is usually smaller than 0.1% of the cantilever length. Such small deformation makes the air damping the dominant effect when operating under ambient conditions. Since the speed of the cantilever is much smaller than the average speed of the air molecules, the air damping force can be considered to be proportional to the speed of the cantilever.⁵³ Therefore the vibrating cantilever can be considered as a driven damped harmonic oscillator. The equation of motion is

$$m^* \ddot{z} + b \dot{z} + kz = F_0 \cos(\omega_d t), \quad (3.27)$$

where m^* is the effective mass, b is the damping coefficient, k is the spring constant of the cantilever, F_0 is the amplitude of the driving force, and ω_d is the angular frequency of the periodic driving force.

The displacement of the cantilever is $z = A(\omega_d) \cos(\omega_d t - \delta)$. The amplitude of the cantilever vibration is

$$A(\omega_d) = \frac{A_0}{\sqrt{\left(1 - \left(\frac{\omega_d}{\omega_0}\right)^2\right)^2 + \frac{1}{(\omega_0 \tau)^2} \left(\frac{\omega_d}{\omega_0}\right)^2}}, \quad (3.28)$$

where $A_0 = A(0) = \frac{F_0}{k}$ is the drive amplitude, $\omega_0 = \sqrt{\frac{k}{m^*}}$ is the natural frequency of the cantilever, and $\tau = \frac{m^*}{b}$ is the relaxation time. The phase angle is

$$\delta = \arctan\left(\frac{\omega_d / \tau}{\omega_0^2 - \omega_d^2}\right). \quad (3.29)$$

The resonance spectra represented by equation (3.28) are shown in Figure 3.7 with three sets of parameters.

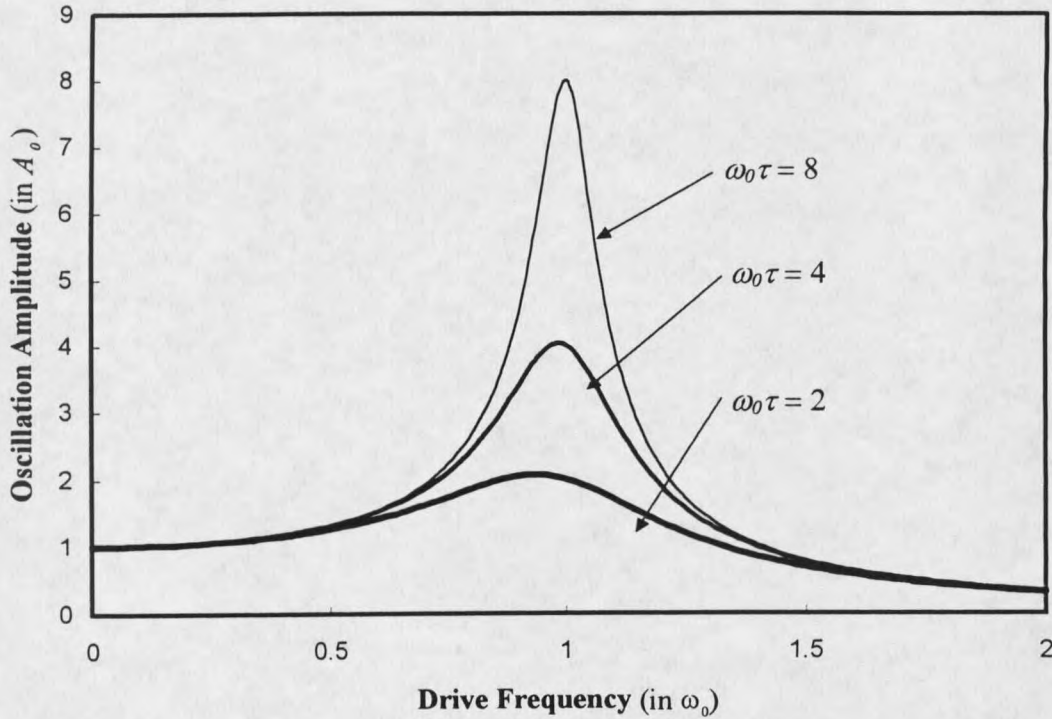


Figure 3.7. The resonance spectra of a driven damped harmonic oscillator.

One of the important parameters for a damped harmonic oscillator is the quality factor, Q . It is defined as 2π times the ratio of the energy stored to the average energy loss per cycle,⁵⁴ that is,

$$Q = 2\pi \frac{\text{energy stored}}{\langle \text{energy loss per cycle} \rangle}. \quad (3.30)$$

When the cantilever is a lightly damped system, that is $\tau \gg 1/\omega_0$, the quality factor of the system can be expressed as⁵⁴

$$Q = \omega_0 \tau. \quad (3.31)$$

For silicon cantilevers that are used for the vibrating cantilever mode, the Q factors are generally greater than 300 under ambient conditions.

From equation (3.28) and the condition $dA/d\omega_d = 0$, we have the resonant frequency,

$$\omega_R = \sqrt{\omega_0^2 - \frac{1}{2\tau^2}} = \omega_0 \sqrt{1 - \frac{1}{2Q^2}}, \quad (3.32)$$

where the oscillation amplitude reach its maximum value. Combining equations (3.28) and (3.32), we have the maximum amplitude,

$$A(\omega_R) = \frac{A_0 Q}{\sqrt{1 - \frac{1}{4Q^2}}} \cong A_0 Q, \quad (3.33)$$

because most of the cantilevers that we use have $Q > 300$.

Tip-Sample Contact: Harmonic Oscillator with Non-Damping Force

In the AFM vibrating cantilever mode operation, where the cantilever is vibrating perpendicular to the sample surface (see Figure 2.2), the tip-sample interaction force applied on the cantilever is independent of the speed of the cantilever. In the case of viscous damping, the damping force is always in the opposite direction of the movement. However, in the case of tip-sample interaction, the force can be in the same direction of the movement during half of a cycle. This can result in the *increase* of the vibration amplitude. To contrast this to the viscous damping, we call the tip-sample interaction the non-damping force. When the tip-sample gap is small enough, physical contact between the tip and the sample can occur. The non-damping force between the tip and the sample plays an important role in the tip-sample contact process.

The tip-sample contact can be classified into two types based on the kinematic status of the tip prior to contact (Figure 3.8). When the tip-sample interaction force is attractive, its direction is against the direction of the spring force of the cantilever while the probe position is lower than its equilibrium position. When the spring force cannot overcome the attractive tip-sample interaction force, the probe will be accelerated towards the sample until it contacts the sample. Since the probe is pulled closer to contact the sample abruptly, this process is called *jump-to-contact* (JTC). Another type of tip-sample contact is caused by that the trajectory of the oscillating tip intersects with the sample surface. In this type of contact, the spring force is greater than the tip-sample interaction force during the whole approach process, and the probe is decelerating prior to contacting the sample. We call this type of contact process *ramp-to-contact* (RTC).

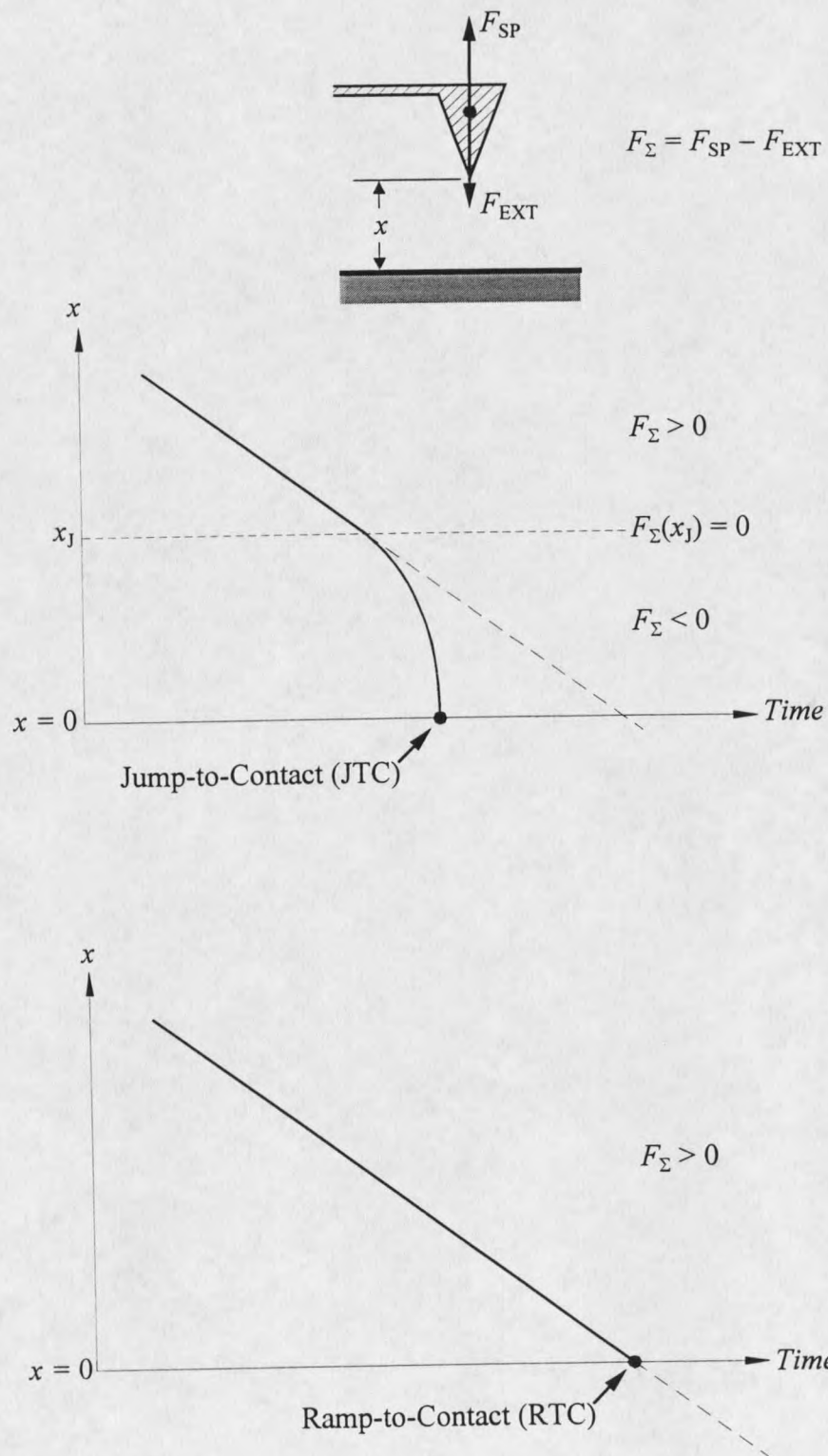


Figure 3.8. Jump-to-contact and ramp-to-contact.

Since the vibration amplitude of a cantilever is usually much smaller than its length, the vibrating cantilever can be considered as a simple harmonic oscillator. In the following analysis, we will use the properties of a simple harmonic oscillator to gain perspective on the conditions that JTC and/or RTC events can occur and the energy transfer when the probe and sample make physical contact during the contact processes.

To perform the following calculation, a simplified model of tip-sample interaction is used, based on the assumption that the capillary force is dominant (Figure 3.9a). The important characteristic is that the force is attractive and its magnitude increases as the tip-sample spacing, x , decreases. Since $dF/dx > 0$, we define this range as having a *positive force gradient*.

If the spring constant of the cantilever is k_0 , and the equilibrium position is x_0 , we have the spring force,

$$F_{SP}(x) = -k_0(x - x_0). \quad (3.34)$$

For the theoretical model of capillary interaction, we assume that the attractive force has a constant force gradient k_1 , the sample surface is at $x = 0$, and the attractive force starts to appear at x_1 . Then, the non-damping force that is shown in Figure 3.9(a) can be written as,

$$F_{EXT}(x) = \begin{cases} 0 & x > x_1 \\ k_1(x - x_1) & x \leq x_1 \end{cases} \quad (3.35)$$

Combining equations (3.34) and (3.35), we have the total force,

$$F_{\Sigma}(x) = F_{SP}(x) + F_{EXT}(x) = \begin{cases} -k_0x + k_0x_0 & x > x_1 \\ -(k_0 - k_1)x + (k_0x_0 - k_1x_1) & x \leq x_1 \end{cases} \quad (3.36)$$

From equation (3.36), when the vibrating probe encounters the attractive force with positive force gradient with respect to the tip-sample gap, the effective spring constant is reduced from k_0 to $k_0 - k_1$. This will cause an increase of the probe displacement. Considering the total force for $x = x_1$, we have $F_{\Sigma}(x_1) = k_0(x_0 - x_1) > 0$ for $x_0 > x_1$. When the tip approaches further, the F_{Σ} will decrease, while maintaining repulsive until the tip reaches a possible threshold point, x_1 ($< x_1$), where $F_{\Sigma}(x_1) = 0$. From equation (3.36), we have

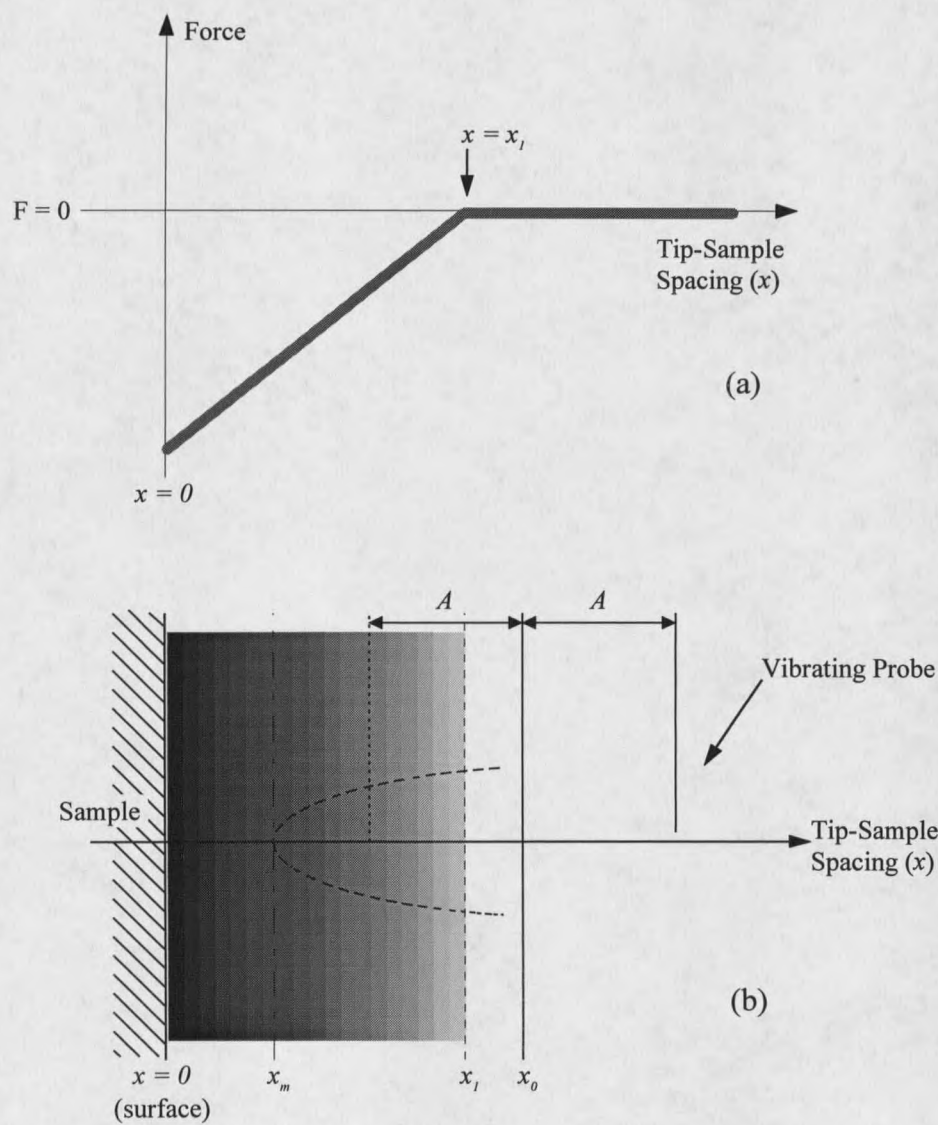


Figure 3.9. A simplified model of a vibrating probe in an capillary interaction with the sample. (a) The force model of the capillary force between the tip and the sample. (b) The tip movement related to the attractive force region.

$$x_J = x_1 \left(\frac{\lambda \mu - 1}{\lambda - 1} \right), \quad (3.37)$$

where $\lambda = k_0/k_1$ is the relative spring constant of the cantilever, and $\mu = x_0/x_1$ is the relative tip-sample gap.

For different λ and μ , the total force, F_Σ , and the possible threshold point, x_J , are plotted in Figure 3.10. When the tip-sample attraction is stronger than the spring force and the equilibrium position is outside of the attractive force region (Figure 3.10a), JTC will occur if $x < x_J$. When the tip-sample attraction is weaker than the spring force and the equilibrium position is outside of the attractive force region (Figure 3.10b), JTC never occurs. When the tip-sample attraction is stronger than the spring force and the equilibrium position is within the attractive force region (Figure 3.10c), JTC always occurs. When the tip-sample attraction is weaker than the spring force and the equilibrium position is within the attractive force region (Figure 3.10d), JTC occurs only if $x_J = 0$.

To gain further understanding of JTC, let us study the first case (Figure 3.10a) in more detail. In this case when $x > x_J$, $F_\Sigma > 0$, which damps the cantilever vibration. However, when $x < x_J$, $F_\Sigma < 0$. The direction of the total force is the same as the direction of cantilever displacement, which breaks the balance and results in JTC. The parameter x_J is the threshold of JTC. For JTC to occur in reality, we must have $x_J > 0$, that is,

$$\lambda = \frac{k_1}{k_0} > \frac{x_0}{x_1}, \quad (3.38)$$

and

$$\lambda = \frac{k_1}{k_0} > 1, \quad (3.39)$$

which are the conditions of the spring constants under which the JTC would happen. From the conditions (3.38) and (3.39), we should increase k_0 (that is, use stiffer cantilevers) to prevent JTC. The conditions of JTC with various λ and μ values are summarized in Figure 3.11.

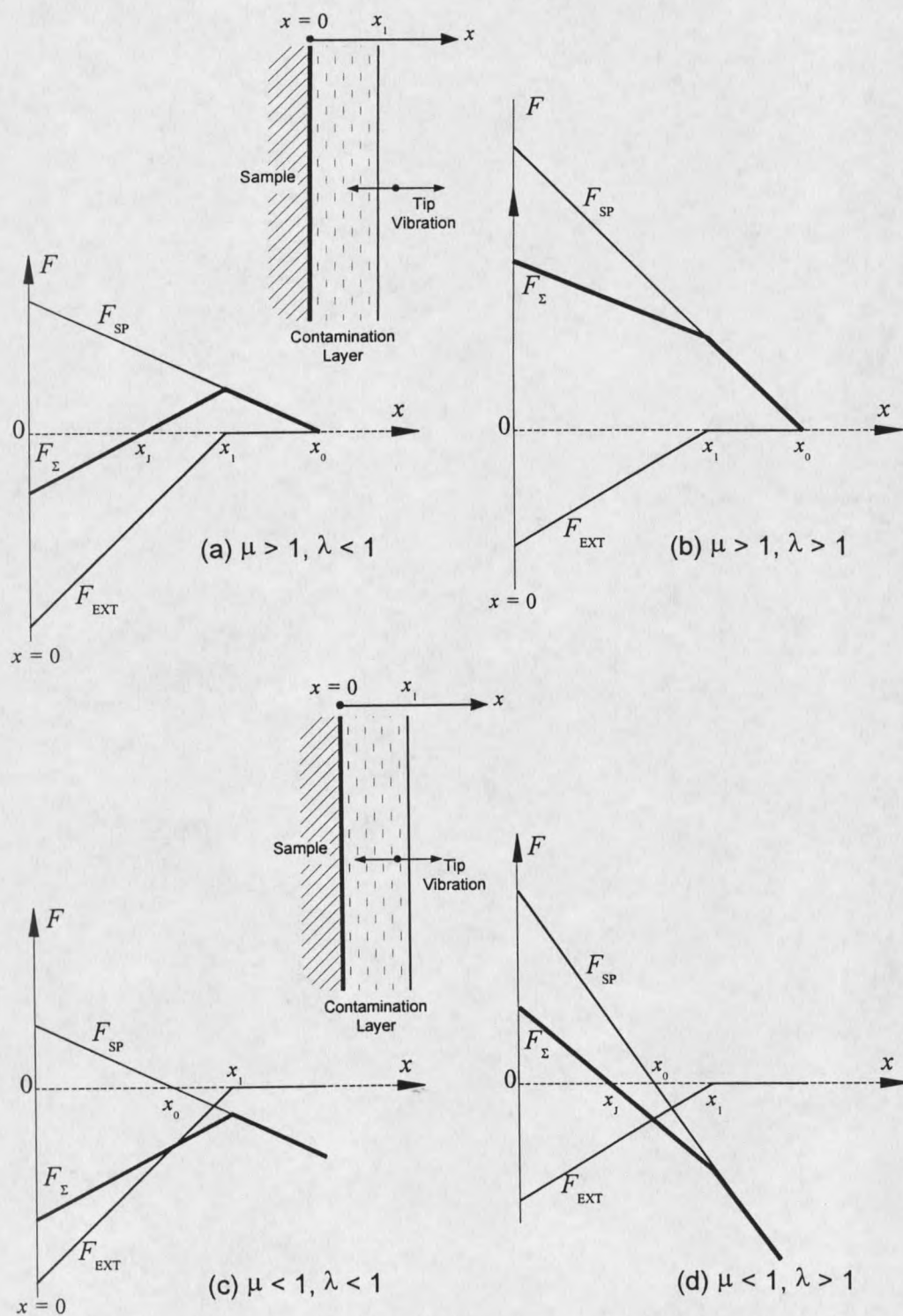


Figure 3.10. The total force applied to the AFM probe and the conditions of JTC: (a) JTC occurs when $x < x_J$; (b) JTC never occurs; (c) JTC always occurs; (d) JTC occurs only for $x_J = 0$.

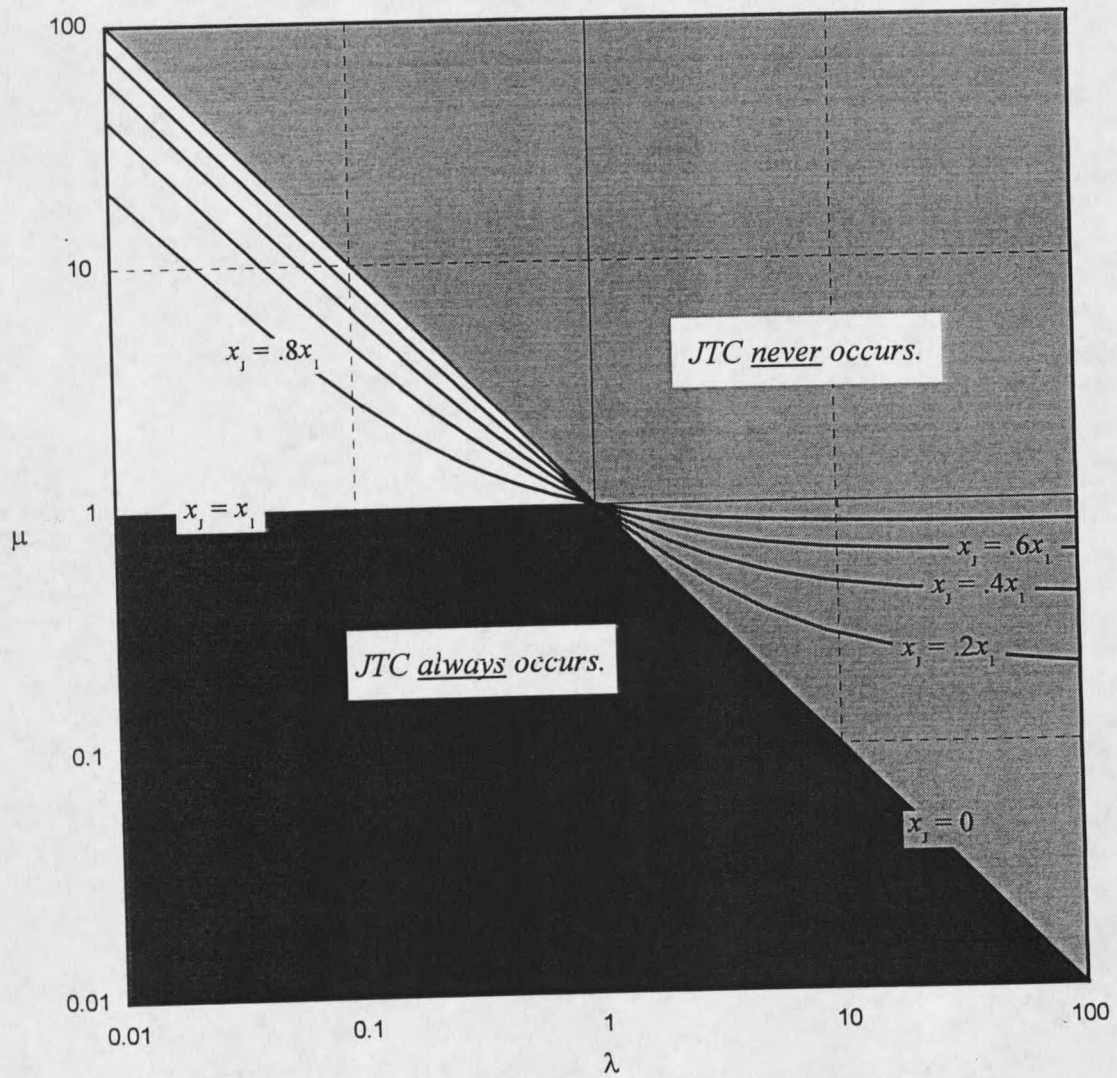


Figure 3.11. Conditions of JTC as a function of the relative tip-sample gap μ and the relative force constant λ .

To determine the condition of RTC, it is important to calculate the lowest point, x_m (see Figure 3.9b), that the probe attains when entering the attractive force region. Figure 3.9b shows the relation between the vibrating probe and the attractive force region. Suppose A is the free oscillation amplitude, then the total energy of this vibrating cantilever is $E = (1/2)k_0A^2$. Since we are studying the situation that the probe is entering the attractive force region, we have $x_0 > x_1$ and $A > x_0 - x_1$. The total potential energy at the cantilever's lowest position, x_m , is

$$\begin{aligned}
 V(x_m) &= \int_{x_m}^{x_0} [F_{SP}(x) + F_{EXT}(x)] dx \\
 &= \int_{x_m}^{x_1} [-k_0(x - x_0) + k_1(x - x_1)] dx - \int_{x_1}^{x_0} k_0(x - x_0) dx \\
 &= \frac{1}{2}(k_0 - k_1)x_m^2 - (k_0x_0 - k_1x_1)x_m + \frac{1}{2}(k_0x_0^2 - k_1x_1^2). \quad (3.40)
 \end{aligned}$$

At the lowest position the potential energy equals the total energy. This gives an equation for x_m ,

$$(k_0 - k_1)x_m^2 - 2(k_0x_0 - k_1x_1)x_m + k_0x_0^2 - k_1x_1^2 - k_0A^2 = 0. \quad (3.41)$$

That is,

$$\begin{aligned}
 x_m &= \frac{(\lambda x_1 - x_0) + \sqrt{(\lambda x_1 - x_0)^2 - (\lambda - 1)(A^2 + \lambda x_1^2 - x_0^2)}}{\lambda - 1} \\
 &= x_J + \frac{\sqrt{(\lambda x_1 - x_0)^2 - (\lambda - 1)(A^2 + \lambda x_1^2 - x_0^2)}}{\lambda - 1}. \quad (3.42)
 \end{aligned}$$

When $x_m < 0$, RTC occurs.

Figure 3.12 shows the calculation results for a special case of $x_0 = 50$ nm, $x_1 = 10$ nm. From Figure 3.12(a), we can see that the downward amplitude increases as λ increases. To reduce the effect of the tip-sample contact it is important to have a small λ value, that is, using a cantilever with greater spring constant, k_0 . Figure 3.12 (b) shows the relation of this downward overshoot with the free vibration amplitude, A .

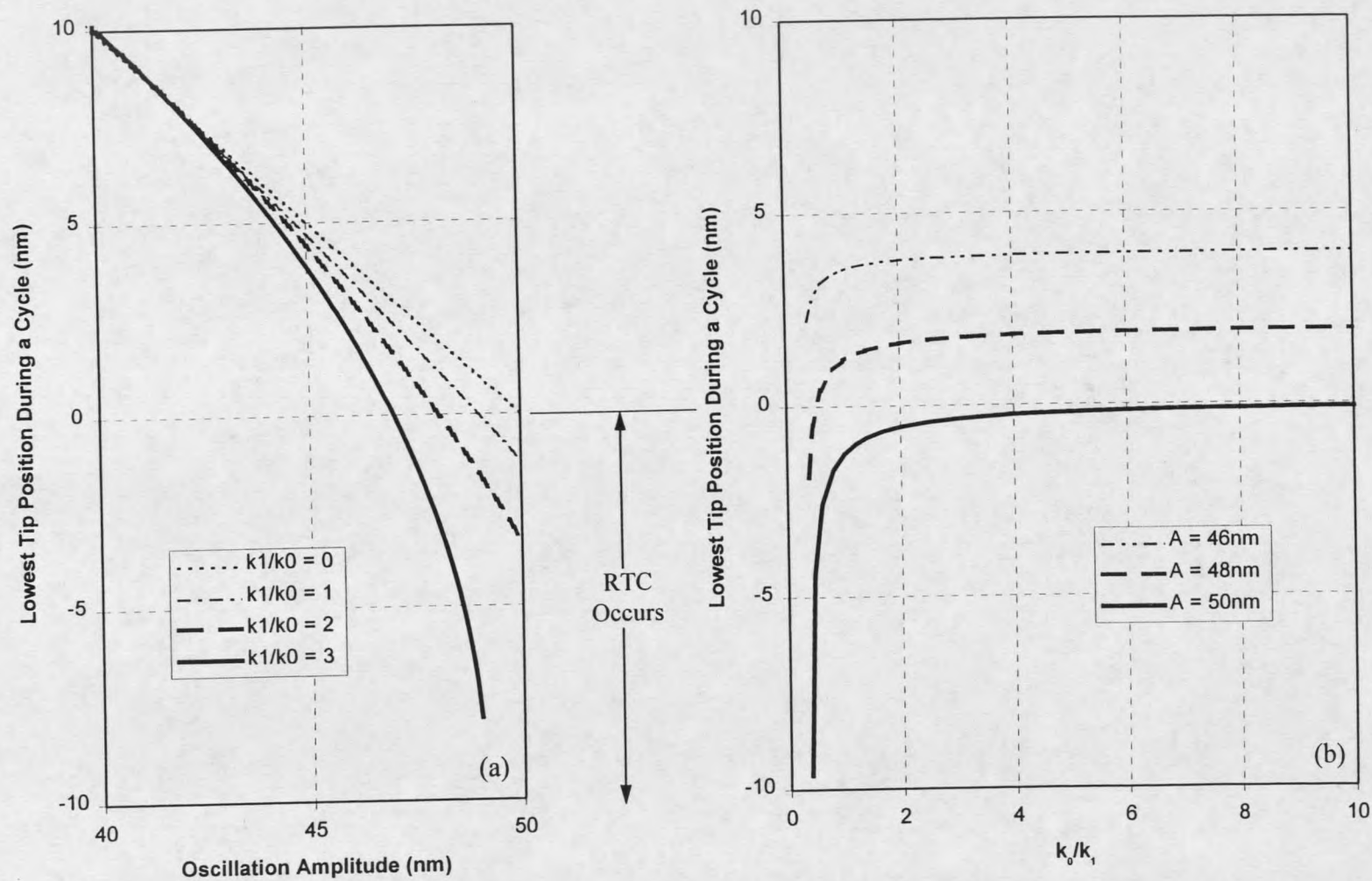


Figure 3.12. Simple Harmonic Oscillator under an attractive force between the tip and sample. In this example, $x_0 = 50\text{ nm}$, $x_1 = 10\text{ nm}$.

It is clear that the attractive interaction with a positive force gradient between the tip and the sample will only increase the vibration amplitude. The amplitude reduction in the vibration has to be caused by the interaction with a negative force gradient. Suppose the sample is a hard surface at $x = 0$; the cantilever will stop at this point. With this assumption, we can calculate the energy transfer at the surface,

$$E_{TRF} = \frac{1}{2}k_0A^2 - V(0). \quad (3.43)$$

Because

$$V(0) = \frac{1}{2}(k_0x_0^2 - k_1x_1^2), \quad (3.44)$$

we have

$$E_{TRF} = \frac{1}{2}k_0(A^2 - x_0^2 + \lambda x_1^2). \quad (3.45)$$

Figure 3.13 shows the calculation result for $x_0 = 20$ nm and $k_0 = 50$ N/m. The amount of energy transfer represents the intensity of tip-sample contact. To minimize the energy loss due to tip-sample contact, the operation amplitude should be small. From Figure 3.13, we can see that cantilevers with a greater spring constant can be operated at larger amplitude without energy loss.

Surface Electrical Potential on Semiconductors

For semiconductors used in industry, one of the important parameters is dopant concentration. In order to measure bulk dopant concentration, one method is to cleave the specimen, and to analyze the changes in surface electrical properties due to the variation of dopant concentration. A good example of the variation of dopant concentration is the p-n junction. Figure 3.14(a) and (b) shows a simple model of a sharp p-n junction.

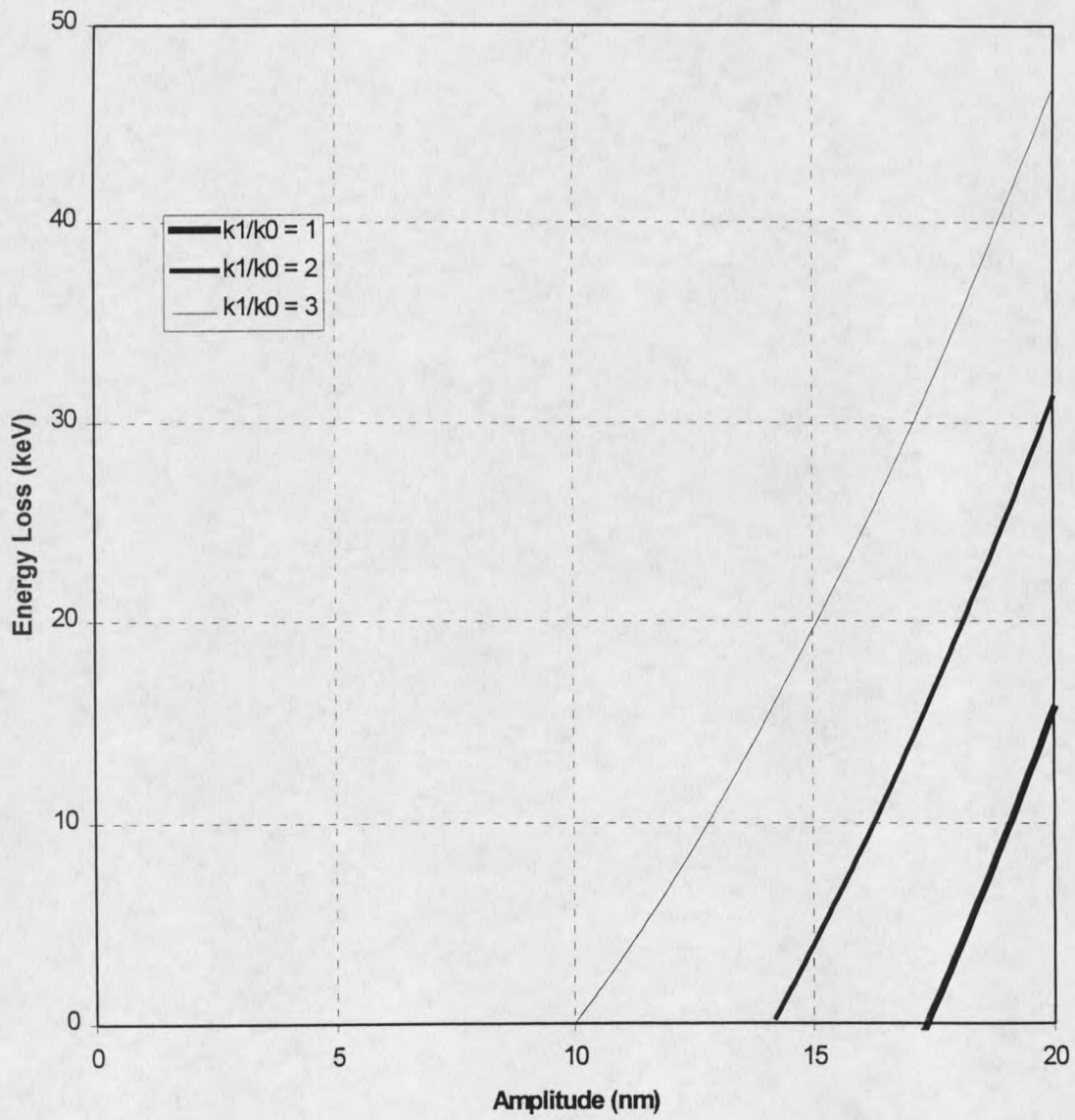


Figure 3.13. Energy loss when the tip encounters a rigid surface.

Due to the difference in their work function between the p-doped region and n-doped region, charges will be transferred across the interface in a semiconductor p-n junction. Eventually the Fermi level on both sides line up and band bending will occur near the interface. The charge concentration near the interface will produce a significant change in the potential. Figure 3.14(c) shows the change of the electric potential across the interface that is given by⁵⁵

$$\phi(x) = \begin{cases} \frac{eN_A}{2\epsilon\epsilon_0}(x + w_p)^2 & -w_p < x < 0 \\ \Delta\phi_0 - \frac{eN_D}{2\epsilon\epsilon_0}(x - w_n)^2 & 0 < x < w_n \end{cases} \quad (3.46)$$

where x is the coordinate perpendicular to the interface, N_A is the acceptor concentration, N_D is the donor concentration, $\Delta\phi_0$ is the difference of the work function, w_p and w_n are the widths of the depletion layer on each side of the p-n junction.

From the above formula, the dopant concentration can be measured by measuring the change of electrical potential across the interface. One method is to cut the sample to expose the cross-section of the interface. When the surface is uniform and free of localized defects, the measured surface potential on the cross-section should be proportional to $\phi(x)$.

With the Kelvin probe force microscopy, the surface potential change across a junction can be measured, which is a measurement of $\phi(x)$. From equation (3.46), the measurement of the surface potential can be a method of measuring the dopant concentration N_A and N_D .

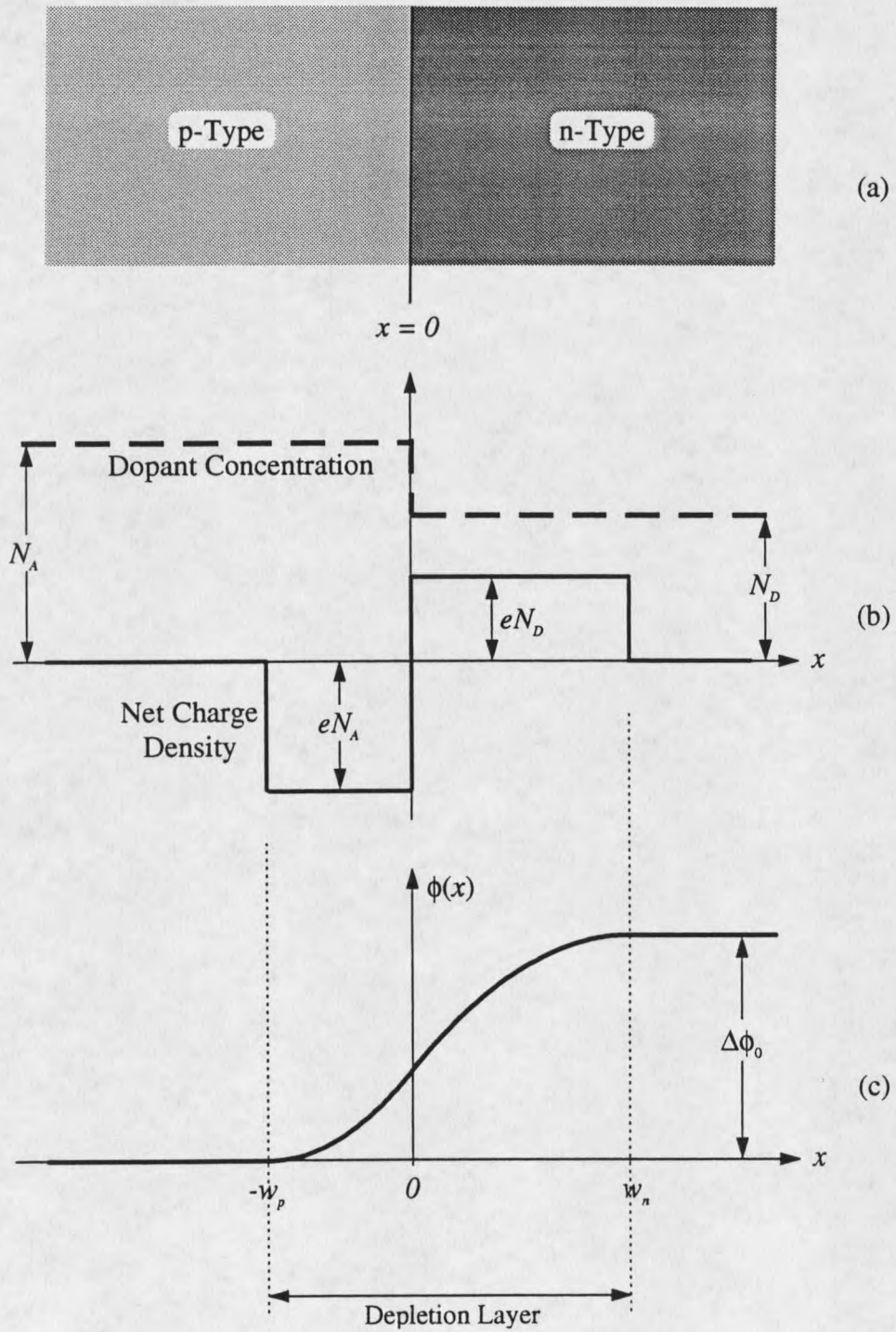


Figure 3.14. (a) Diagram of a sharp p-n junction;⁴⁰ (b) Dopant concentration and net charge density; (c) Electrical potential.

Summary

The theoretical analysis presented here is mostly related to nanometer scale surface interactions. Much of the content is taken from previously published materials. This thesis presents three new subjects of theoretical discussion. The first is the derivation of the final formula of the capillary force based on the constant liquid volume assumption. The parameter G_0 in the final formula, equation (3.18), clearly gives a meaning to the thickness of the surface liquid layer. The second is the definition of the theoretical lateral resolution of PPM. The third is the introduction of a new concept, ramp-to-contact (RTC), which has significant differences from the conventional jump-to-contact (JTC), and the theoretical analysis that leads to the establishment of the conditions in which JTC and RTC occur.

CHAPTER 4

EXPERIMENTAL APPARATUS

Introduction

To perform experiments that are able to study tip-sample interactions at nanometer scale, a complex instrumental arrangement is needed. A new PPM system was designed and built to operate simultaneously with a scanning electron microscope. The system allows the PPM tip to be positioned precisely over a sub-micrometer scale feature. Next, a feedback control technique, which is the vibrating cantilever mode (VCM), was used to maintain a gap at nanometer scale between the tip and the sample. To measure the electrical potential changes, an additional feedback loop was built based on the traditional Kelvin method. During the research of this thesis, three types of high aspect ratio AFM probes were used.

Design of the Combined PPM/SEM System

An important area of application of PPM is the semiconductor industry. The sizes of the electronic devices are getting smaller each year. By the year 2007, the minimum feature size is expected to reach 0.1 micrometer.⁵⁶ In order to locate the device of interest to inspect with a PPM, a proper ranging technique must be used. In the past, the optical microscope was the only practical ranging tool. When the feature size is smaller than one micrometer, which is beyond the resolving power of the optical microscope, the PPM itself must be used to scan a larger area and zoom in to locate a small feature. This would significantly increase the imaging time. When the sample is po-

rous and not guided by any ranging tool, the PPM will fail because the tip is most likely to get into a hole and not able to engage a surface. The solution is to develop a PPM with the scanning electron microscope as its ranging tool.

Design Objectives of the Instrument

For a PPM/SEM system to be practical, the following requirements must be met. It must be able to perform both STM and AFM imaging. For the AFM, both the continuous contact and vibrating cantilever modes must be available. The optical deflection detection method can meet this requirement. It should be able to operate simultaneously with a SEM. To perform electrical measurements, both the tip and the sample bias must be electrically controllable.

Figure 4.1 shows the basic configuration of the instrument. To place the area of interest under the PPM probe, two levels of translation stages are needed. First, the secondary translation stage moves the PPM probe to the center of the SEM field of view. Then, the primary translation stage moves the sample under the PPM probe, so that the area of interest can be located on the sample surface. The tip must be directly visible by the SEM to allow accurate positioning of the tip to sub-micrometer areas of interest; that is, view of the tip must not be blocked.

To increase operational efficiency, both the tip and the sample should be replaceable without breaking the vacuum of the main SEM chamber. Since the tip can be damaged in various continuous contact or periodic contact modes,⁵⁷ it is important to monitor the condition of the probe tip before and after PPM operation with the SEM. To achieve angstrom scale resolution, a vibration-isolation system must be integrated into the combined PPM/SEM system. A new PPM/SEM system was designed and built by the author of this thesis that completely fulfilled the above requirements.¹¹

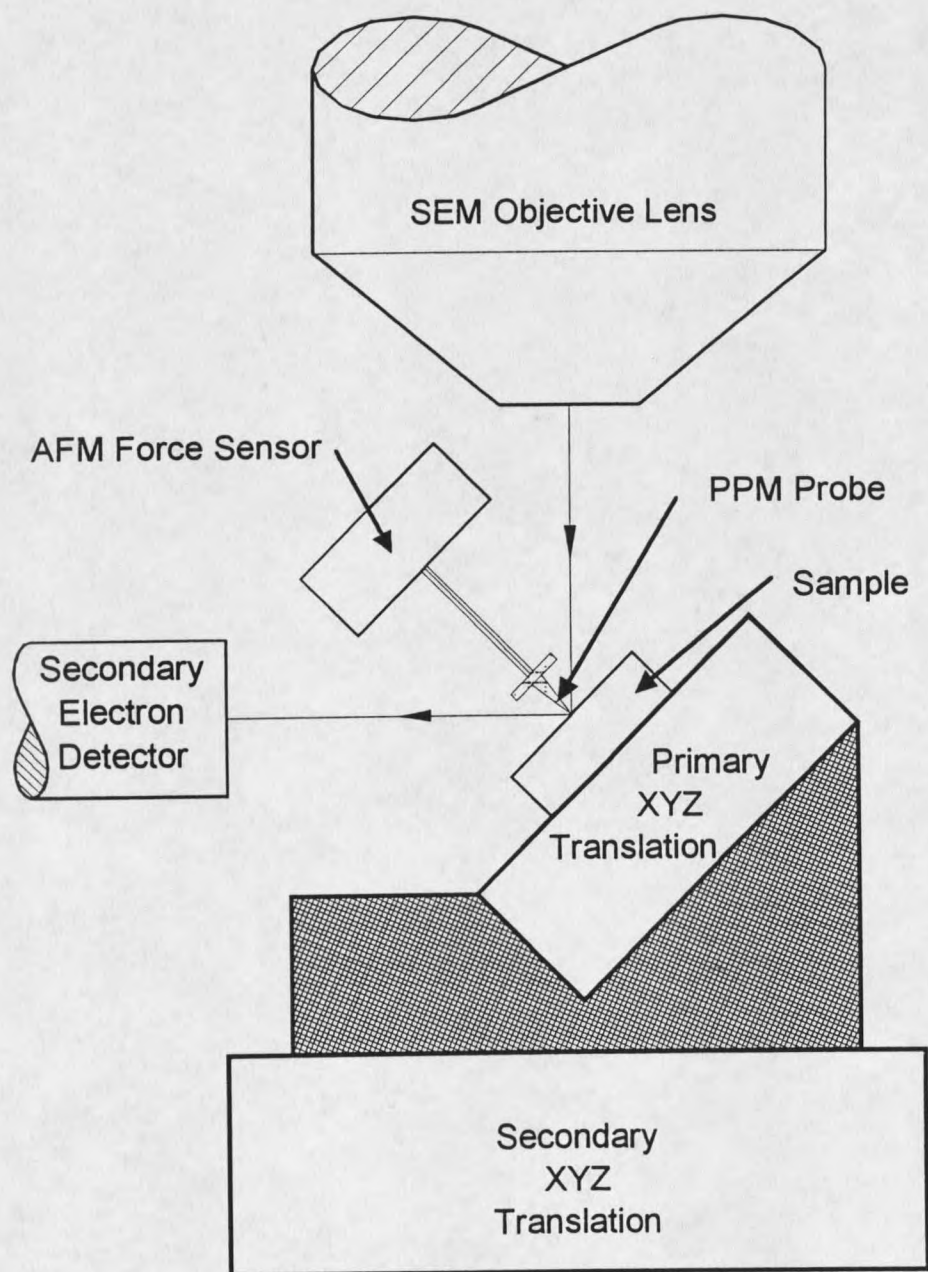


Figure 4.1. Configuration diagram of the PPM/SEM combination system.

Overview of the Instrument

This new PPM was built on a Hitachi S-4100 FE-SEM. The original sample stage of the SEM was removed and the PPM stage was then installed on the same port. Figure 4.2 is a diagram showing the PPM mounted to the SEM specimen chamber, while Figure 4.3 shows a photograph of this instrument pulled out of the vacuum chamber.

The PPM is built on a rectangular flange, which in turn is mounted to the SEM's specimen chamber. On the vacuum side, a framework is built to hold the PPM stage, which supports the PPM head. The framework is also designed to provide clearance for the electron gun, the secondary electron detector, and the energy dispersive X-ray (EDX) analyzer.

A load-lock chamber is built on the atmospheric side of the rectangular flange, with the primary purpose of enabling fast tip and sample replacement without breaking the vacuum of the SEM specimen chamber. The SEM specimen chamber is separated from the load-lock chamber by a gate valve through which the whole PPM head can be moved to the load-lock chamber. After introducing air into the load-lock chamber through an exhaust valve, the cover of the load-lock chamber can then be pulled open to provide access to the PPM head. In this position, tip, sample and scanners can be replaced, and the AFM's laser can be aligned.

To allow optimal SEM viewing of both the sample and the tip of the PPM probe simultaneously, the sample is placed at 45° relative to the electron beam, and the PPM probe is perpendicular to the sample surface. This arrangement can be seen in Figure 4.1. To position the PPM tip and sample to their operating positions requires two stages of translation. The secondary translation moves the PPM head relative to the electron beam, and the primary translation moves the sample relative to the PPM tip.

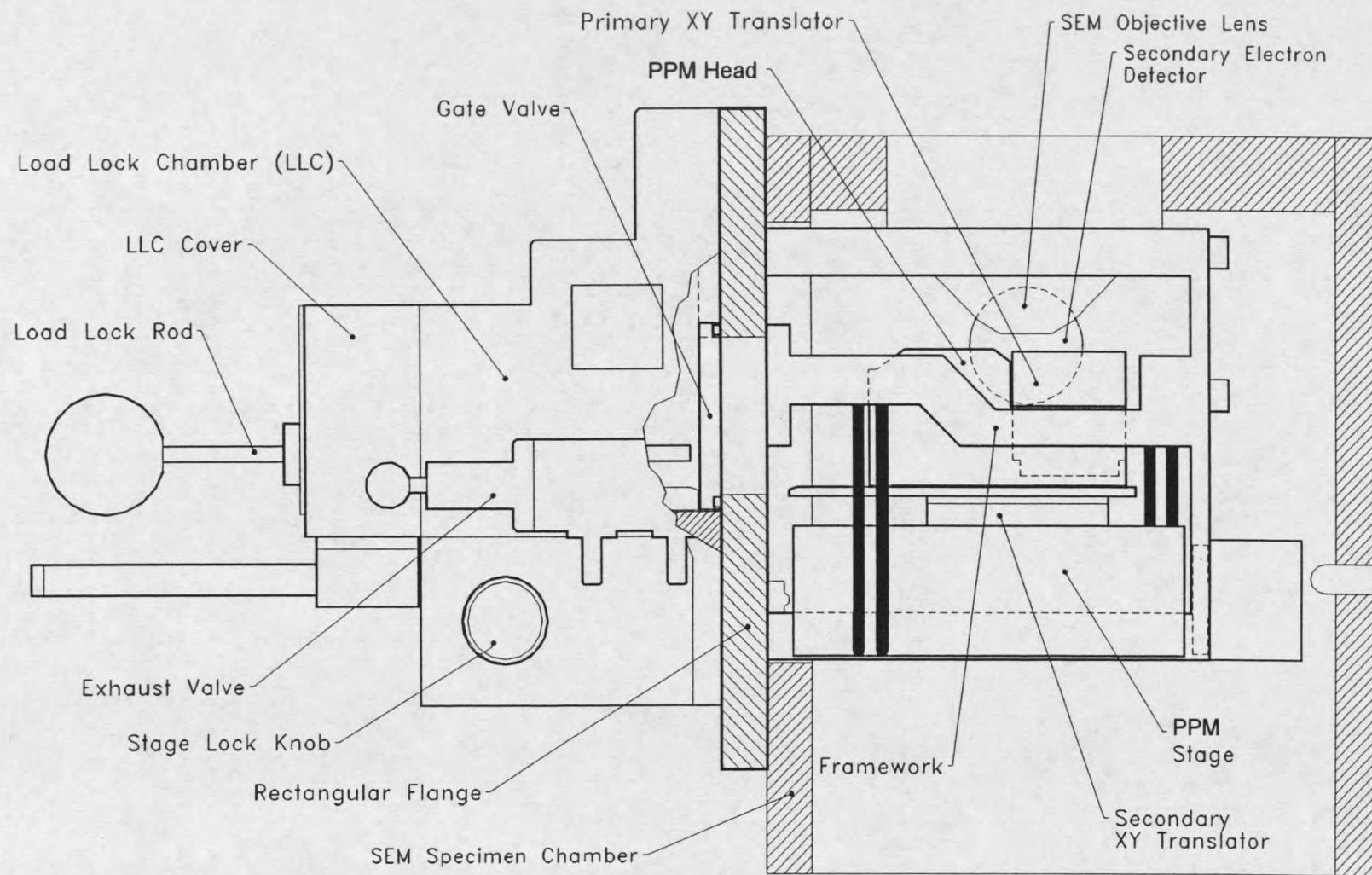


Figure 4.2. Diagram of the PPM integrated with a scanning electron microscope.

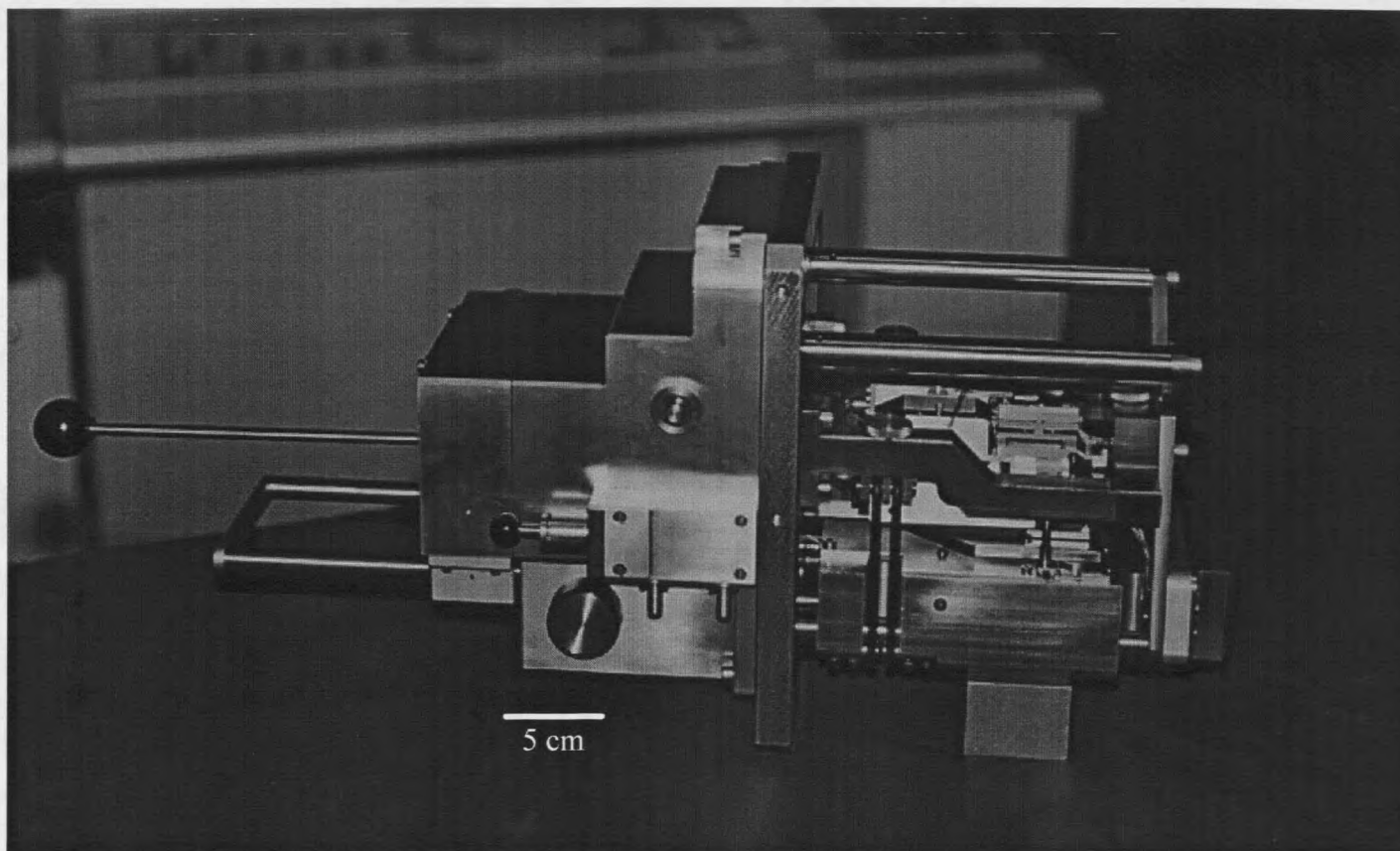


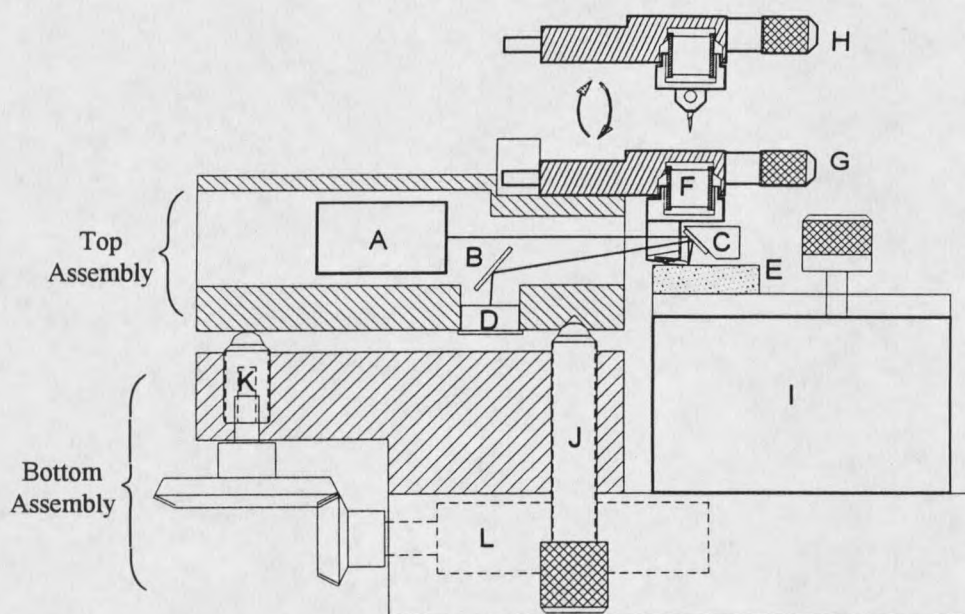
Figure 4.3. Picture of the PPM unit removed from the SEM specimen chamber.

PPM Head

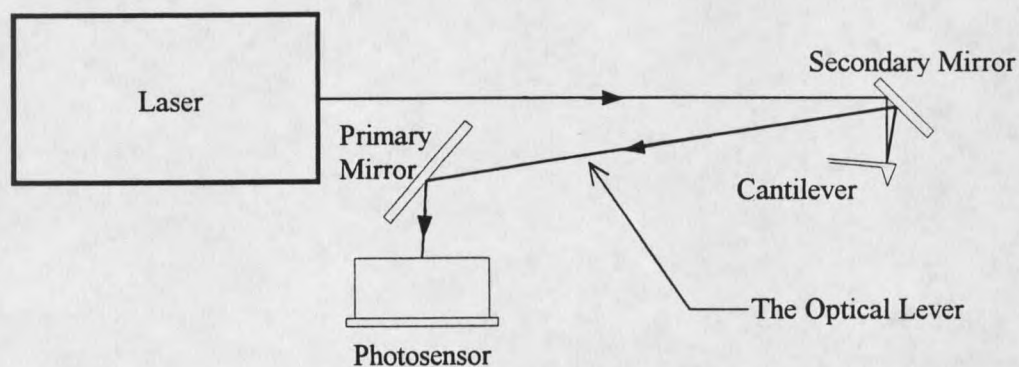
Figure 4.4 shows the basic structure of the PPM head, which consists of the scanner (STM or AFM), the AFM force sensor, the sample translator (the primary XY), and tip-sample gap control (the primary Z). The PPM head can perform either STM or AFM imaging. Both STM and AFM pre-amps are built into the PPM head. In order to switch between STM and AFM, only the scanner needs to be exchanged.

The AFM force sensor is of the laser deflection detection type.⁵⁸ The laser system that is built into the PPM head consists of a semiconductor diode laser, the primary deflection mirror with controllable angle, the secondary deflection mirror with fixed angle, and the photosensor. The angle of the primary deflection mirror is controlled by a DC motor and a pair of limit switches. The secondary mirror deflects the laser beam to and from the cantilever, for the purpose of deflecting the laser optics away from the electron beam, allowing the SEM viewing of the PPM tip. This design allows us to work at reasonably short SEM working distances. Figure 4.4(b) shows the geometry of the AFM laser deflection detection optics. The length of the optical lever is 36.5 mm.

The PPM head consists of two major assemblies, the top assembly and the bottom assembly (Figure 4.4a). The PPM probe is mounted to the top assembly and the sample is mounted to the bottom assembly. The top assembly is supported by the bottom assembly through three fine-pitch screws (J and K) that are used to adjust the spacing between these two assemblies. Since the PPM head is held at 45° relative to gravity, the top assembly and bottom assembly of the PPM head are held together by four heavy force springs. Changing the spacing between these two assemblies changes the tip-sample gap, which is controlled by two manual thumbscrews (J) for coarse approach and one motorized screw (K) for fine approach. An optical eye loupe can be placed on the PPM head to monitor the coarse tip approach. In the fine approach, the top assembly of the PPM head pivots on the thumbscrews (J). The direction of the linear movement of the motorized screw is opposite to the movement of the tip. The reduction ratio of the tip approach, which is the ratio of the linear speed of the motorized screw to the speed of the tip approaching the sample, is 2.8 to 1.



(a) The design of the PPM head.



(b) The AFM laser deflection detection optics.

Figure 4.4. The PPM head. (A) laser diode, (B) deflection mirror, (C) secondary mirror, (D) photoelectric sensor, (E) Sample, (F) piezo tube, (G) AFM scanner, (H) STM scanner, (I) primary XY translator, (J) manual adjustment screw, (K) motorized screw, (L) DC motor.

Both circuit boards for the STM and AFM pre-amps are built into the PPM head. The short distance between the pre-amps and the probe helps to reduce electrical noise. This arrangement also makes it easy to perform AFM topography and electrical conductivity imaging simultaneously. When performing surface electrical conductivity imaging, the metal-coated AFM probe is electrically connected to the input end of the STM pre-amp to measure the current flow between the tip and the sample.

Piezoelectric Scanners

In a PPM, a piezoelectric scanner is used to create a raster scanning motion between the tip and the sample. The scanner uses piezoelectric ceramics to convert electrical voltage to mechanical motion. (The term piezoelectric ceramic is usually abbreviated to *piezo*.) In this system, the scanner is of the single tube scanner design, which uses a piece of tube-shape piezo; a design commonly used in PPM.⁵⁹ The reason for using the tube scanner is its compact size and small scan range for high-resolution imaging. In this system, the tip is scanned and the sample is fixed. In the AFM scanner, the cantilever is scanned while the laser system maintained stationary.⁶⁰ The scan range is limited to less than 6 μm to keep the majority of the laser spot maintained on the cantilever. The advantage of the tip-scanning technique is that the relationship between the scan plane and sample surface is controllable.⁶¹ The piezo is completely shielded by the stainless steel base and a stainless steel cover to prevent the voltage changes on the piezo from distorting SEM's electron beam. These two metal parts (base and cover) are electrically connected by a spring washer made of beryllium copper. The probe holder for AFM cantilevers or STM tips is situated on the top cover. The probe holder is electrically isolated from the metal shell by a layer of Torr Seal[®]. On the AFM scanner, a cantilever is mounted by a spring clip. A bias voltage can be applied to the cantilever for scanning Kelvin-probe force microscopy (KFM) and electrical conductivity imaging. The STM scanner includes a metal tube to mount the STM tip which is made of a tungsten wire.

Sample and Probe Replacement

Samples, probes, and scanners can be replaced without breaking the vacuum of the SEM specimen chamber. In this scanning-tip design, the sample is mounted on top of the primary translator and the probe is mounted on the scanner, which is plugged into the top assembly of the PPM head. The movable PPM head can be operated in either the main chamber position or the load-lock chamber position.

Both the main chamber and the load-lock chamber are equipped with docking stations. When the PPM head is in the SEM specimen chamber, it is mounted on top of the microscope stage with a kinematic design.⁶² When the PPM head is in the load-lock chamber, it rests on a sliding platform. It is transferred along the sliding platform between the two chambers by a load-lock rod.

The PPM head can be operated in either chamber. The electrical cables that drive the PPM are connected to both chambers, with an internal electrical connection system built into both the PPM stage and the load-lock chamber. While being transferred from the load-lock chamber onto the PPM stage, the PPM head is automatically disconnected from the electrical cable on the load-lock chamber, and automatically connected to the electrical cable on the stage.

Vibration Isolation System

To guarantee sub-angstrom resolution, a rigid foundation is needed for optimal vibration isolation. A rigid framework for supporting the PPM on a set of Viton O-rings is built on the vacuum side of the rectangular flange (see Figure 4.5). The PPM stage is based on a stainless steel mass plate, which in turn is suspended from the framework by six Viton O-rings. The O-rings are stretched vertically with their upper ends hooked to the middle bar of the framework and their lower ends hooked to the bottom of the mass plate (see Figure 4.5). Together with the rigid foundation, this suspension system provides excellent vibration isolation for sub-angstrom resolution imaging.

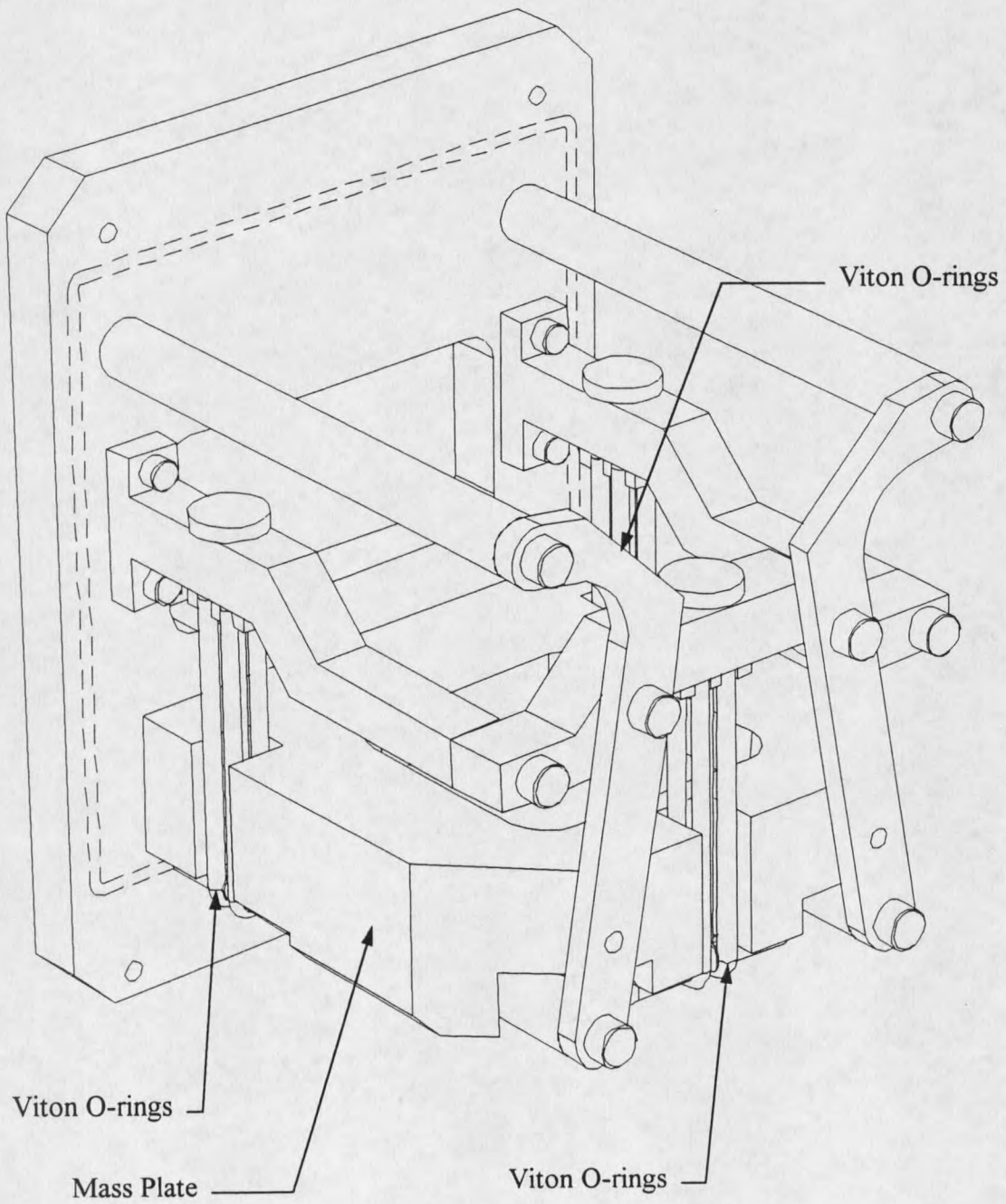


Figure 4.5. The vibration isolation system.

For SEM imaging, the PPM stage must be locked down to remove any sample movement relative to the electron beam. A locking mechanism built on the framework locks the stage by clamping the mass plate horizontally with a pair of stainless steel clamps. The movement of the clamps is controlled by turning a knob on the outside of the load-lock chamber.

Translation Stages

There are two levels of translation in this PPM. The primary X-Y-Z translation system is built on the PPM head. The primary XY translator moves the sample under the PPM probe while the primary Z translation controls the tip-sample gap. The secondary X-Y-Z translation system is built on the PPM stage. The secondary XY moves the whole PPM head horizontally relative to the electron beam. This allows the PPM tip to be moved to the center of the field of view of the SEM. The secondary Z translation adjusts the working distance of the SEM. In normal operation, the PPM probe is first moved to the center of SEM view by the secondary translation. Then the area of interest on the sample is moved to the PPM probe by the primary XY stage.

The primary XY translator is of the inertial piezoelectric drive type.⁶³ It has nanometer scale translation steps, extremely high translation speed, zero backlash and no jerking. This translator is also rigid enough for sub-angstrom scale imaging. The XY travel range of this translator is 6.3×6.3 mm. The primary Z motion is driven by a DC motor through a set of 1:2 miter gears. The Z travel range is 6 mm.

The secondary XY translation moves the whole PPM head, and was tested to be able to carry a load as large as 20 kg. Each axis of this translator is driven by a DC motor, with the travel range of 11 mm and minimum step size of about $0.1 \mu\text{m}$. Since the field-of view of the SEM can be shifted in a range of $\pm 10 \mu\text{m}$, the PPM tip can be moved to the center of the SEM's field-of-view easily.

The secondary Z translation is mainly used to reduce the working distance for the SEM in order to increase the SEM resolution. The range of working distance for the Hitachi S-4100 type

SEM is between 14 mm and 25 mm. The secondary Z motion is generated by three fine-pitch screws. They are synchronized by a drive train that is able to move a load of 6 kg at a speed of 33 mm/min. A built-in photoelectric position sensor monitors this translation, preventing the PPM from crashing into the SEM objective lens and allowing the PPM stage to return precisely to the load-lock position for transferring the PPM head.

Control System

This PPM is controlled through a TopoMetrix electronic control unit (ECU) and a specially designed electronic control system. Figure 4.6 show the schematic of the electronic control system.

The PPM stage contains both motion generation and motion sensing devices. It includes two sets of XY and Z motions and a motorized primary mirror rotation for the AFM. The primary mirror rotation and the secondary Z motion have motion-sensing devices associated with them.

When working in vacuum, it is difficult to know the angular position of the AFM primary mirror, which is rotated by a DC motor. A set of mirror rotation limit sensors, consisting of two electrodes, one on each end of the mirror's rotation, was built into the PPM head. When the limit is reached, the electrode is grounded, providing the limits of the mirror rotation. The secondary Z position is also hard to determine when the system is inside the SEM chamber. Therefore, a photoelectric position sensor is built on the PPM stage to monitor the secondary Z position. With this sensor, the PPM stage can be moved precisely to the load-lock position.

Direct Probe Viewing

Since the PPM probe is at 45° relative to the electron beam, the tip condition and tip-sample interaction can be monitored with the SEM. For AFM probes, in order for the probe tip to be visible, the length of the probe must be greater than half the cantilever width, or the shape of the end of the cantilever needs to be redesigned so that it does not block the probe tip view. This tip-viewing requirement is achieved by the VLS silicon probe.

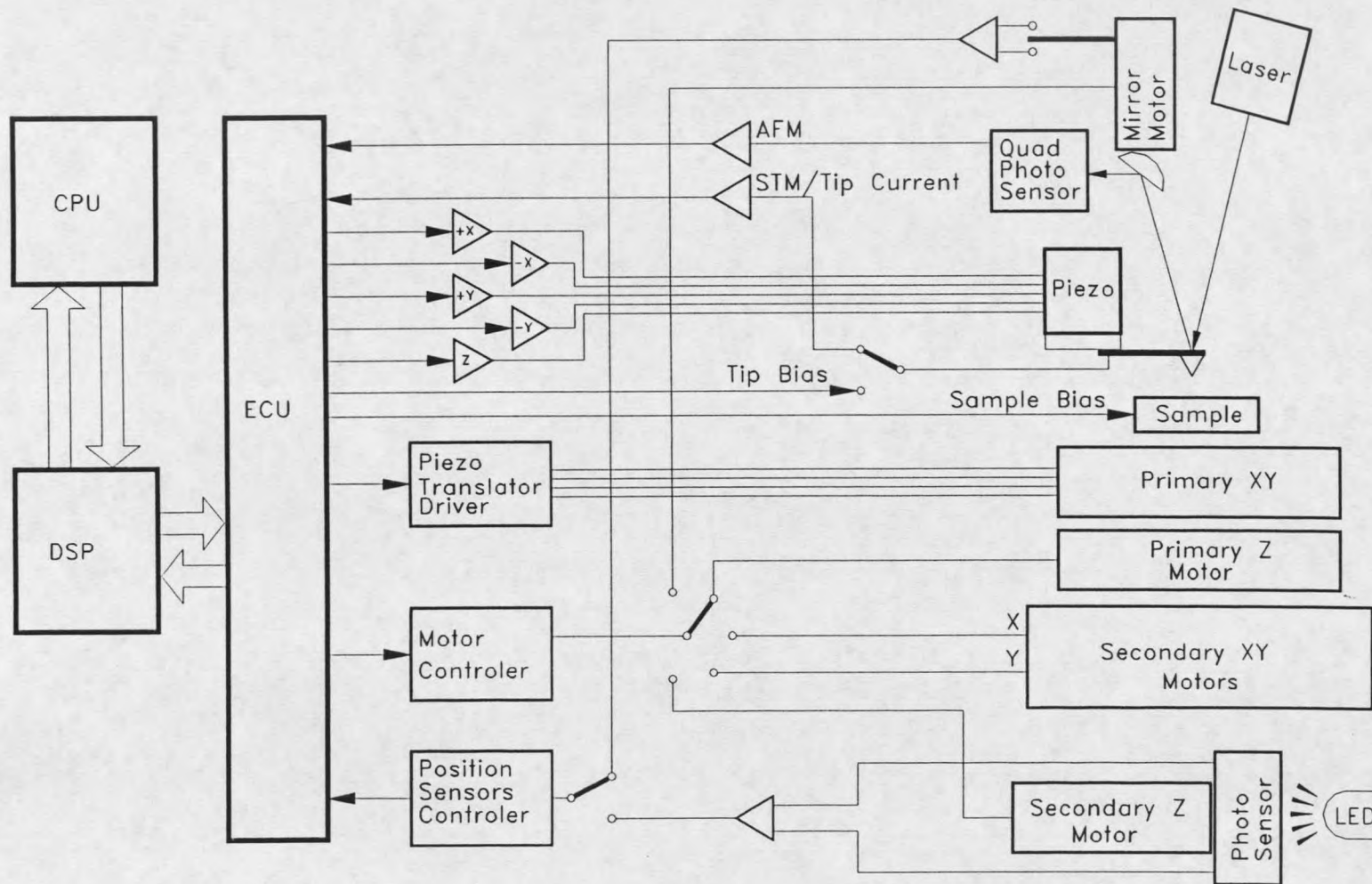


Figure 4.6. Schematic diagram of the electronic control system.

Besides viewing the tip at 45° , another tip viewing position is built into the PPM head. From the normal working position, a PPM probe can be rotated to a horizontal position, where the tip is perpendicular to the e -beam. In this position, the sharpness of the tip can be easily inspected. Figure 4.7 shows two examples of different tip conditions of silicon probes.

Testing the Performance of the PPM/SEM System

Operation in Vacuum

The most critical issue in vacuum operation is the change in the photoelectric detector signal when moving the PPM into vacuum. The signal changes depend on whether the PPM head is in air or in vacuum. The change in the photoelectric detector signal is most obvious during the initial chamber evacuation, and indicates that some extra cantilever bending occurs.

There are two major causes of the extra cantilever bending. The first cause is the temperature change of the cantilever, because the cantilever always undergoes some heating from the laser beam. Since heat dissipation in vacuum is much lower than in air, the cantilever will heat up to a higher temperature. For silicon nitride cantilevers, the top side is metal coated. Because of the bi-material structure of this kind of cantilever, different temperatures generate different degrees of cantilever bending. For single-crystal silicon cantilevers, this effect is not noticeable.

The second cause is surface charging, that occurs during the evacuation, which produces an extra electrostatic attraction force onto the cantilever. This mainly occurs on insulating or semi-conducting samples, because the conductivity of these materials is not large enough to neutralize surface charges in a short time. It affects both silicon nitride and silicon cantilevers. The charge-induced bending occurs gradually, indicating gradual charge build-up. After a long time, the charges are neutralized and the cantilever deflection is reduced. The static attraction can make the cantilever move 10 to $100\mu\text{m}$. If the tip is too close to the surface, this could cause severe tip damage when the tip crashes into the sample.

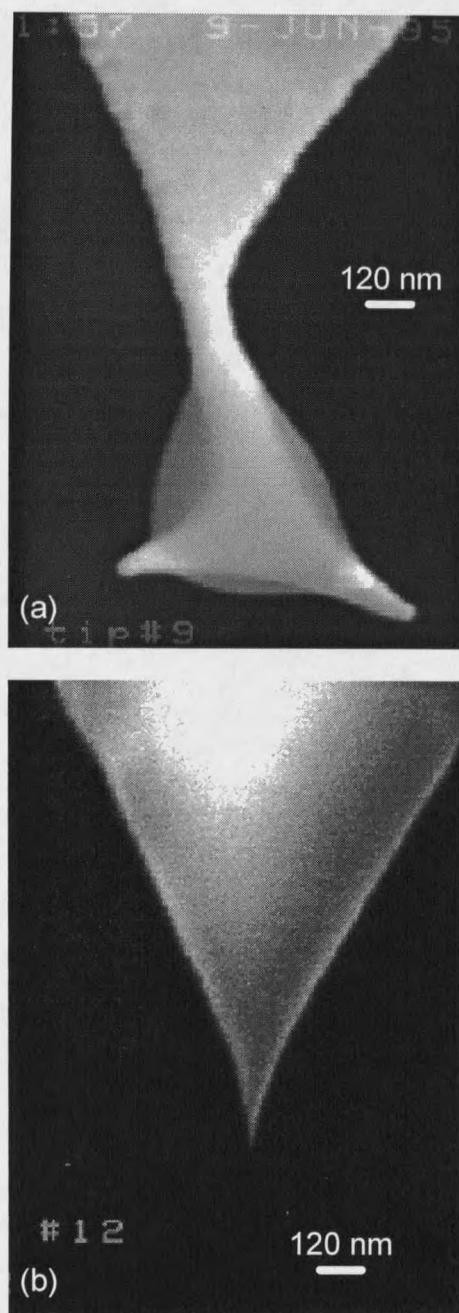


Figure 4.7. Examples of different tip conditions of a type of commercially available silicon AFM tip. (a) A tip that was not etched completely. (b) A regular sharp tip.

Working in vacuum also brought up the potential problem of overheating the laser diode. When the temperature of the laser diode increases, its output power drops dramatically,⁶⁴ and could be reduced to the point that the AFM is not able to function. After improving the heat dissipation of the laser diode, the reduction of the laser total intensity was measured to be less than 3% over 14 hours of continuous operation in vacuum.

Simultaneous PPM/SEM Operation

Since SEM interference on the AFM is generally small, these two microscopes can be operated simultaneously. The laser of the AFM does not affect the SEM's operation. However, it was observed that the electron beam injects 60 Hz noise into an AFM image when the SEM was zoomed-in to the cantilever so that the SEM's field of view was completely blocked by the back side of the cantilever. In such a high SEM magnification (~10k), with the AFM cantilever covering the entire SEM's field of view, it was also observed that an additional force was applied to the cantilever when either the SEM's acceleration voltage or magnification was changed. Further research is needed to understand the mechanism of this electron-beam effect.

On the other hand, the electron beam of the SEM can create major problems for the STM. The hydrocarbon contamination on the sample surface, which is induced by the electron beam,⁶⁵ can greatly affect the tunneling current. When the STM is in operation, the feedback electronics controls the Z piezoelectric ceramic so as to keep the tunneling current at its set point, but the contamination induced by the electron beam can greatly reduce the tunneling current. When this occurs, the feedback electronics, in an attempt to maintain a constant tunneling current, can drive the STM tip into the sample and crash the tip. However, if the contamination is minimized, then it is possible to operate the STM normally with the SEM. In general, to minimize electron beam-induced contamination, the SEM should be operated with higher acceleration voltage and lower magnification.⁶⁶ When running this particular STM, the appropriate setting of SEM acceleration

voltage should be more than 10 kV, the magnification of the SEM should be set to less than 1000X, and the imaging time should be minimized.

Aging of Piezoelectric Scanner

It has been observed that the maximum scan range of the tube scanners decreases over time. The cause of this scanner-aging problem is due to the aging effect of the piezoelectric ceramic, which is the reduction of the piezoelectric constant over time. There are two major causes of piezo aging. The first is relaxation, which causes relatively small range reduction and will eventually stop. The second is fatigue, which can cause dramatic range reduction and will get worse as the scanner is used.

To explain piezo relaxation and fatigue, we need to look into the working mechanism of the piezoelectric ceramic, which consists of many micro-crystals called "grains", and the matter among them called "grain boundary". The grain boundary is basically a material defect, which can easily trap unbalanced charges. This is the origin of the "space charge". When the temperature of ceramics is dropped to room temperature in the fabrication process of the ceramics, the ceramics go through a phase transition (from paraelectric phase to ferroelectric phase). In ferroelectric phase, spontaneous polarization appears in each single cell of the grains.⁶⁷ This process causes the first redistribution of the charge. Although all the charges are stable, a large number of unoccupied electronic states, having even lower energy, exists in the ceramics. Therefore, these space charges are "not-so-stable".

After the poling process, each of the micro-grains has its own polarization that can be viewed as a single dipole moment. Each of these dipoles are oriented differently. When a small voltage is applied to a piezoelectric ceramic, only the magnitude of these dipoles changes. When a large voltage is applied, the micro-grains will rotate so their dipoles line-up, as much as they can, with the external field. Usually, the rotation can be restored when the external field is removed. However, if

excessive stress is also applied to the ceramic, microscopic cracks will develop inside the material, which is fatigue.

In general, when a new condition (such as voltage, stress) is applied to the ceramics, internal changes occur in the material. These are, (1) relaxation: The space charges move to a lower energy state under applied voltage or stress; (2) fatigue: Micro-cracks developed under applied voltage or stress. This physical change creates new space charges and also creates new states with lower energy for the space charge.

When a space charge moves from a higher energy state to a lower energy state, it reduces the total polarization of the ceramics. Therefore, the piezoelectric constant is reduced, and scan range is reduced. Because ceramic is an insulator, the movement of charges to their lowest energy states can be very slow.⁶⁸ For the relaxation process, external forces can speed up the process. Therefore, the scan range can reach a stable value quickly. For the fatigue process, since new space charges are generated, external force will make the problem worse.

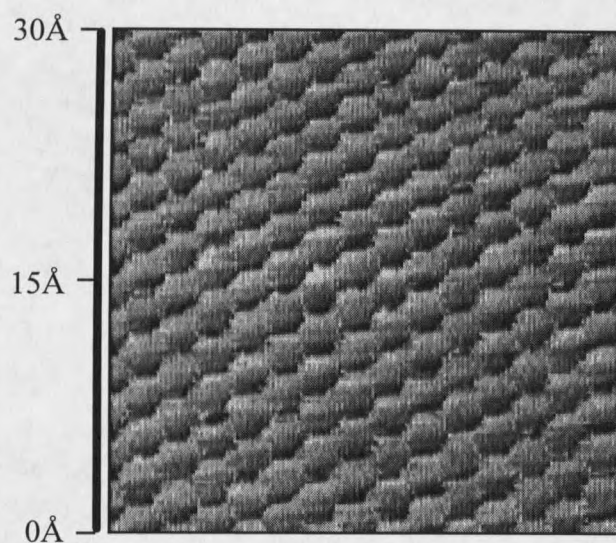
The diameter of the piezoelectric tubes used in this PPM system, D , is 0.25 inch. Two kinds of piezoelectric tubes of different lengths ($L = D$, and $L = 2D$) were used. Compared to the short scanner, the long scanner has a much larger bending angle. The bending angle is $\theta = \sin^{-1}\left(\frac{R}{L}\right)$, where R is the intrinsic scan range and L is the effective piezoelectric ceramic length. Since θ is a small angle, we have, $\theta = \frac{R}{L}$. For the tube-shaped ceramic, the scan range is $R \propto L^2$.⁶⁹ Therefore, we have $\theta \propto L$. For each tube-shaped ceramic, the dead zone on both ends is about 0.03 inch long. The effective length of piezoelectric ceramic for the short scanner is 0.19 inch (4.8 mm). The effective length of piezoelectric ceramic for the long scanner is 0.44 inch (11.2 mm). Therefore, the bending angle of the long scanner is about 2.3 times of the short scanner. The internal stress of the long scanner is much greater than the short scanner.

After one year of operation with maximum operation voltage, the scanners with long piezos showed a range reduction of 20%-40%. The scanners with short piezos showed about a 10% reduction in maximum scan range. If there is only the relaxation process, the range reduction should be similar. The difference in scan range reduction shows that the long piezos had significant fatigue. To ensure the piezoelectric tube has less scan range reduction due to fatigue, a design criteria for selecting the dimensions of the scanning tube is to use a tube with $L \leq D$. If a long tube needs to be used, then the operational voltage must be reduced to minimize the bending angle.

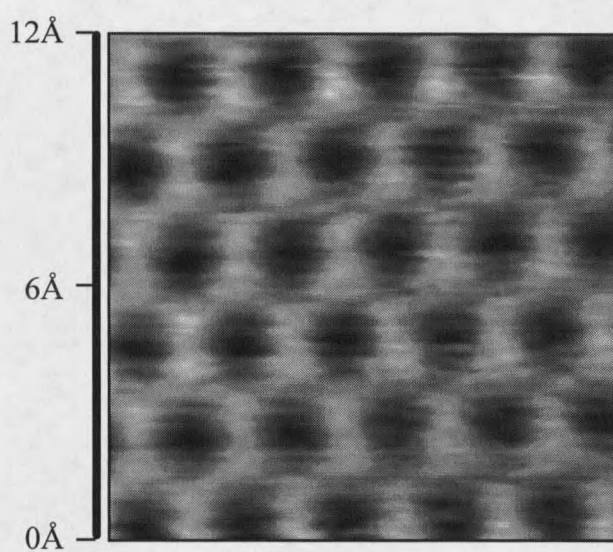
Vibration Isolation and Atomic Resolution

To demonstrate the effectiveness of the built-in vibration isolation system, atomic resolution imaging was tested with highly-oriented-poly-graphite (HOPG). Both AFM and STM images were acquired (Figure 4.8), with imaging time of about 20 seconds. The AFM is performed in the continuous contact mode (no cantilever vibration). Because of the strong tip-sample interaction, the AFM image is not the true atomic image, instead it only contains information of the symmetry of the surface lattice. On the other hand, the STM is capable of achieving single-atom tunneling under ideal conditions, which makes true atomic resolution possible. Since there is no surface contamination existing between the tip and the sample when operating in vacuum, single atom tunneling is possible with a sharp STM tip. True atomic resolution images were acquired on HOPG in vacuum with STM (Figure 4.8b), which shows the well known honey-comb structure unattainable in air.

The system noise spectrum was analyzed in the frequency range of 0 to 2 kHz. The PPM probe was brought to the sample, and the noise spectrum of the photodiode signal or tunneling current was measured (see Figure 4.9). There is no obvious peak in those spectra. This indicated that the system is very stable. The Viton O-ring and heavy mass vibration isolation, along with the vacuum and the SEM chamber, provided a very quiet environment for the PPM, with an rms noise equivalent to 0.02 nm of vibration.



(a)



(b)

Figure 4.8. Images of highly-oriented-poly-graphite (HOPG) acquired in the SEM. (a) AFM force image. The field of view is 30 Å. (b) STM tunneling current image. The field of view is 12 Å. The bias voltage was 50 mV. The tunneling current was 1 nA.

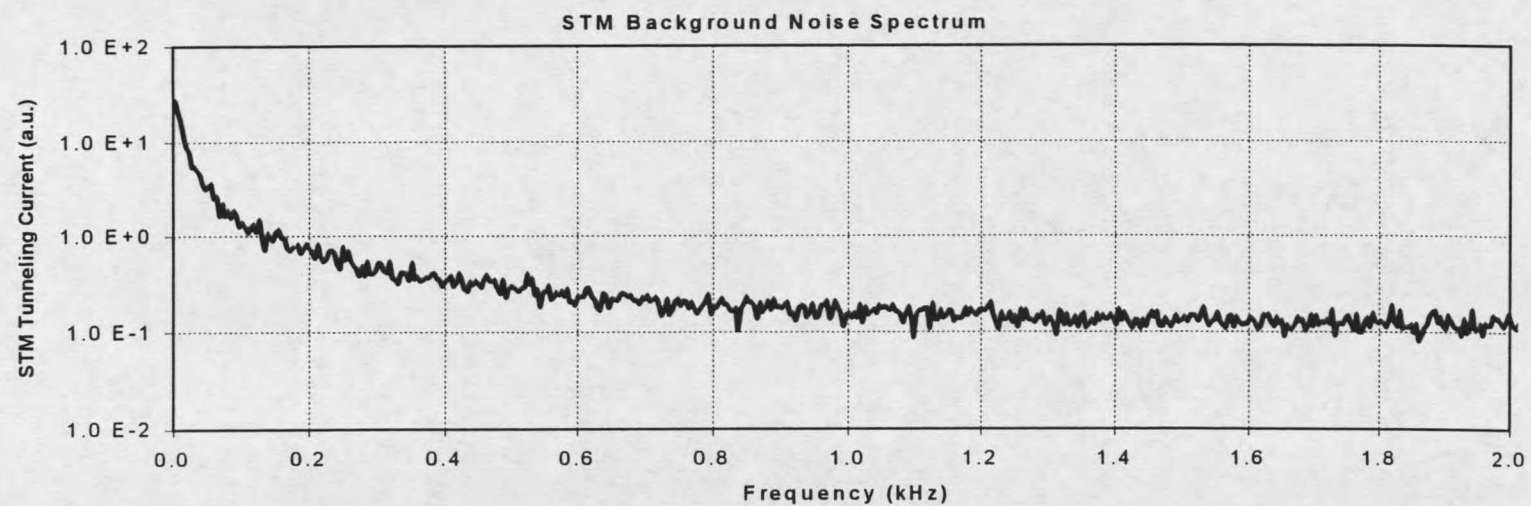
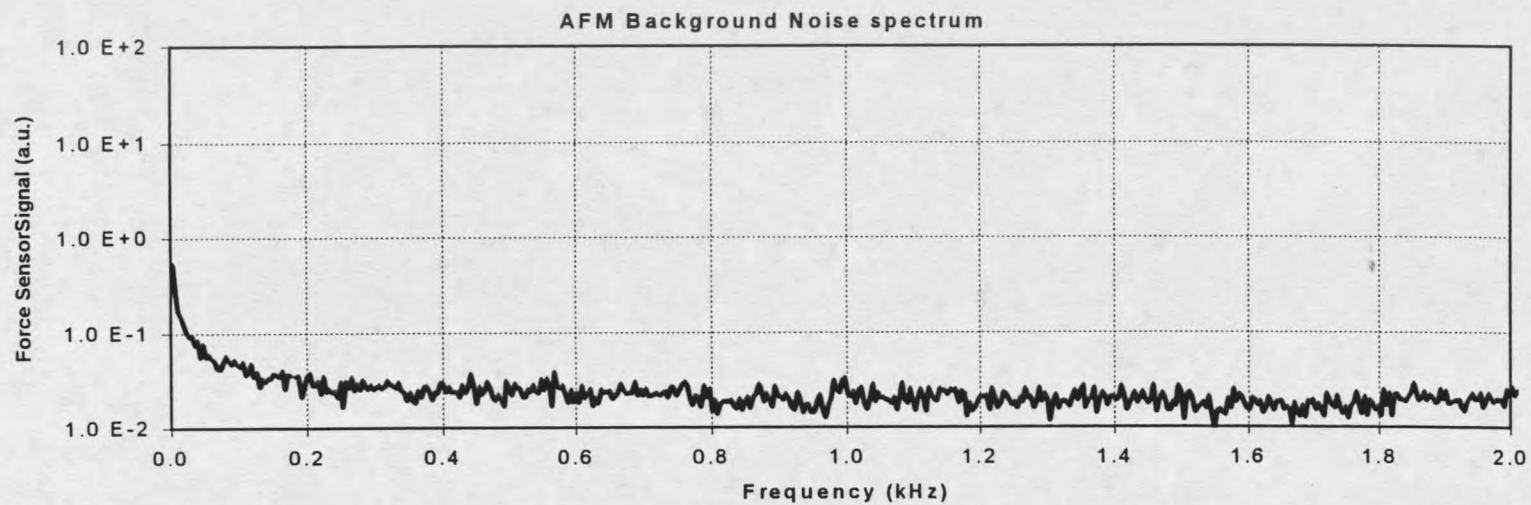


Figure 4.9. The background noise spectra of the feedback signals.

Studies of Tip Conditions

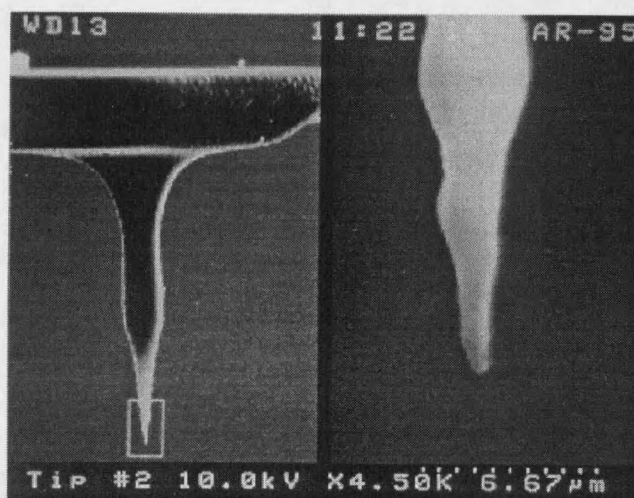
The probe tip condition is very important to the quality and repeatability of PPM imaging. A degraded tip can reduce image resolution and produce imaging artifacts. There have been reports of degraded tips causing changes in surface roughness measurements.⁵⁷ With this instrument, the SEM can be used to monitor the tip condition of a PPM probe, identifying bad tips to help reduce PPM image artifacts.

Figure 4.10 shows the condition of a VLS silicon AFM tip before and after imaging a very rough aluminum grain sample. The near-contact mode AFM image is shown in Figure 4.11. The line profile on the bottom is plotted for X and Z to be the same scale. In this way the slope on the line profile represents the actual slope measured by the AFM tip. From this line profile, we can see that the largest slope of the grains measured is about 70° . From the SEM image, the tip radius is about 20 nm. Although the sample is very rough, no change on the tip was observed before and after the near-contact imaging operation, which gives high reliability to the topographic image shown in Figure 4.11.¹²

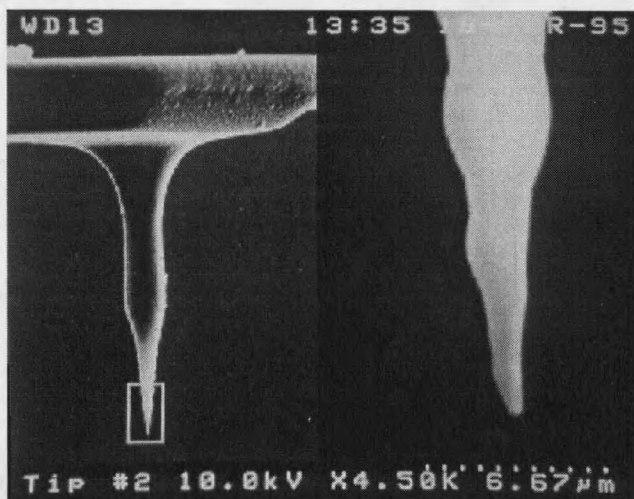
Locating Sub-micrometer Features

To study a feature that is less than one micrometer in size, it is not possible to use an optical microscope. With the AFM, one can always make a large area survey to look for sub-micrometer features. However, this would take a lot of time. With the SEM, one can make use of its high resolution and high scan rate to quickly locate a hard-to-find sub-micrometer feature.

To study the early stage of the development of cracks in a structural ceramic material, a ceramic cross-section sample was imaged, to look for the micro-cracks on the surface, which were about a fraction of a micrometer wide. These micro-cracks were not visible under an optical microscope, and they were not numerous. The only clue was that they might be by the side of a larger crack.



(a)



(b)

Figure 4.10. SEM micrograph of a silicon VLS AFM tip before (a) and after (b) usage. The image on the right hand side is the enlarged tip area of the left hand side image. The increase of the magnification is ten times. The scale bar on the bottom is for the image on the left hand side. For the right hand side image, each division represents 66.7nm.

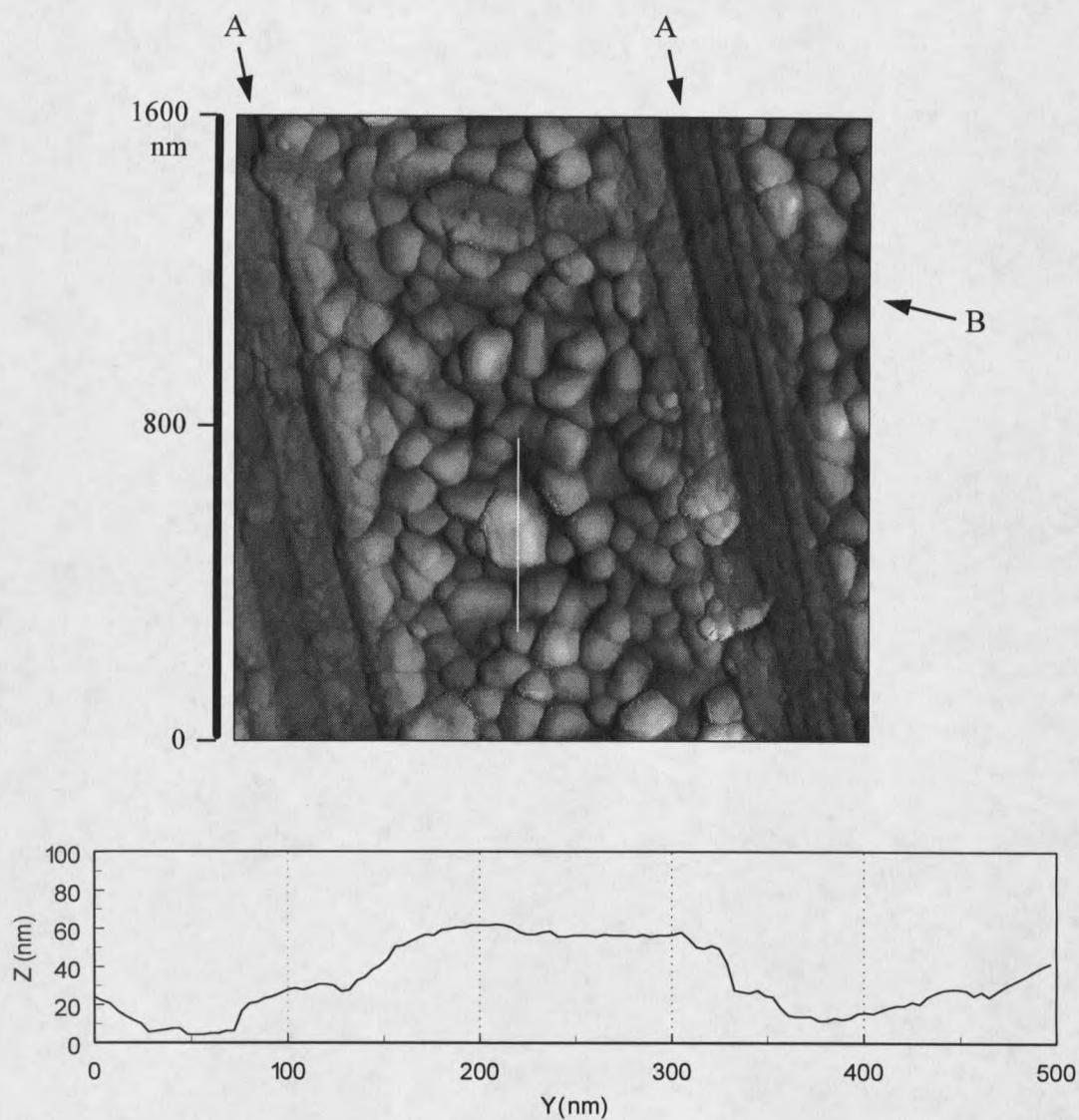


Figure 4.11. Near-contact AFM image of an aluminum grain sample. There are two deep scratch marks (A) and one shallow scratch mark (B) in the image area. The image size is $1.6\text{ }\mu\text{m} \times 1.6\text{ }\mu\text{m}$. The image was acquired in air. The cantilever vibration amplitude was 2 nm.

To study this sample, the AFM tip was moved to the center of the SEM field of view by the secondary XY translator. A large crack was observed on the sample (Figure 4.12(a)). Then the sample was moved by the primary XY translator to search along the crack (Figure 4.12(b)). Before long, a couple of micro-cracks were found (Figure 4.12(c)). A continuous contact mode AFM image was acquired on these micro-cracks (Figure 4.12(d)). The time that was used to complete this sequence is about 20 minutes. In the AFM image, the quarter-micrometer wide crack (A) on the right side can be clearly seen in the SEM image (Figure 4.12(c)). Another smaller crack above this one (B) can also be seen in the SEM image. However, the fine grain structure on the ceramic surface is not visible in the SEM image.

Vibrating Cantilever Mode AFM System

System Structure

In the vibrating cantilever mode (VCM) operation, the AFM cantilever is set into vibration by a modulation voltage applied to the piezoelectric ceramic that supports the cantilever (see Figure 2.4). Normally, the modulation frequency is set near the resonant frequency of the cantilever in order to make use of the high Q-factor of the cantilever to increase force sensitivity. The vibrating cantilever causes the photosensor output signal to oscillate at the same frequency, which is then sent to the input of a phase-locked loop.⁷⁰ Two kinds of output signals can be obtained, the *amplitude* signal and *phase* signal.

The VCM measurements were made with a TopoMetrix modulation unit capable of amplitude and phase detection. Figure 4.13 is a block diagram of the electronics. Figure 4.14 illustrates a typical frequency response of a silicon cantilever used in this paper. The frequency used for scanning was selected from the computer display of the vibration amplitude verses frequency. According to equation (3.33), the vibration amplitude of the probe at resonance, A_m , can be calculated by

$$A_m = Q_p A_e, \quad (4.1)$$

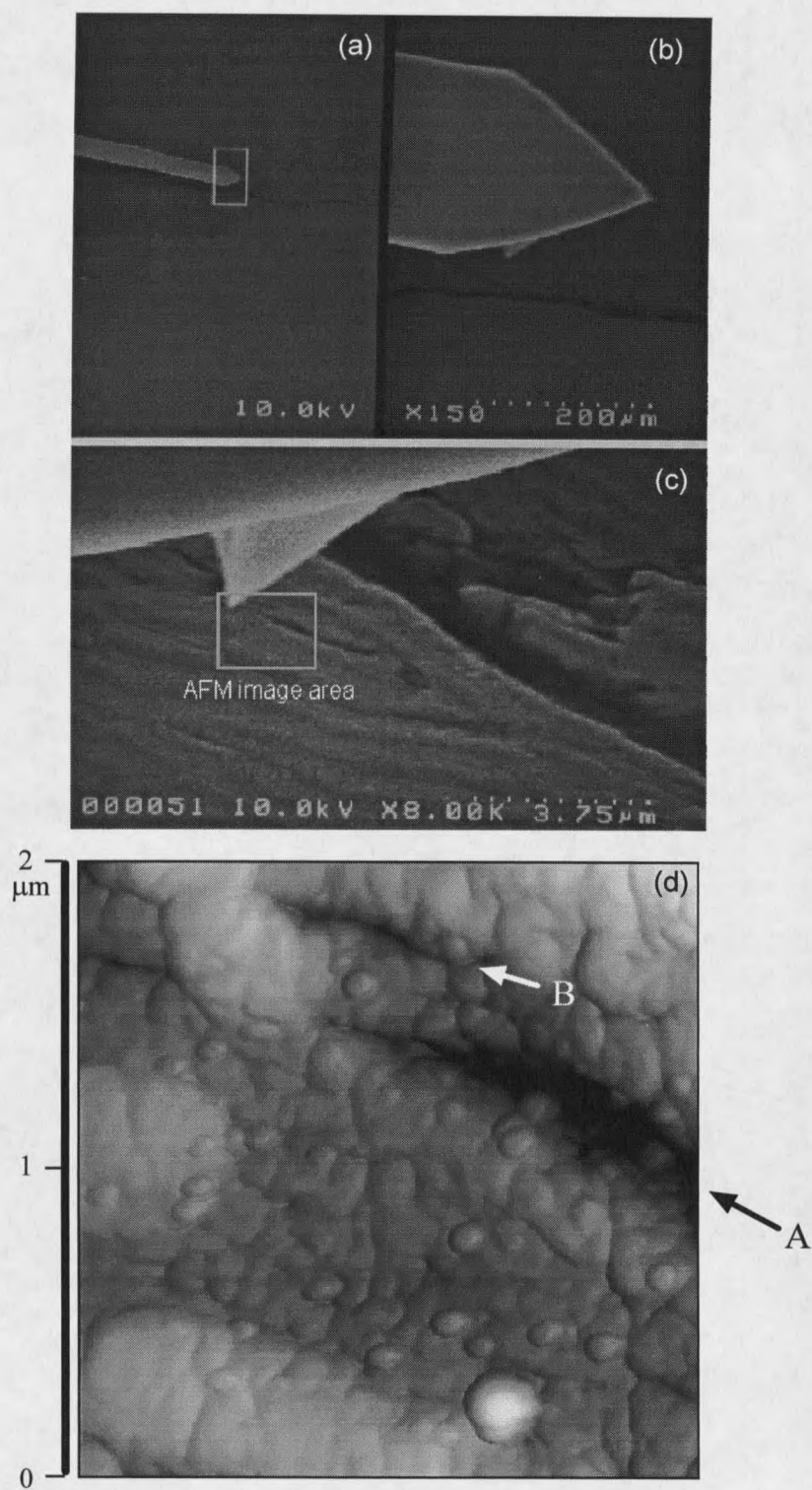


Figure 4.12. Studies of micro crack in ceramic. (a-c) SEM images; (d) an AFM image. The size of the AFM topographic image is $2 \times 2 \mu\text{m}$.

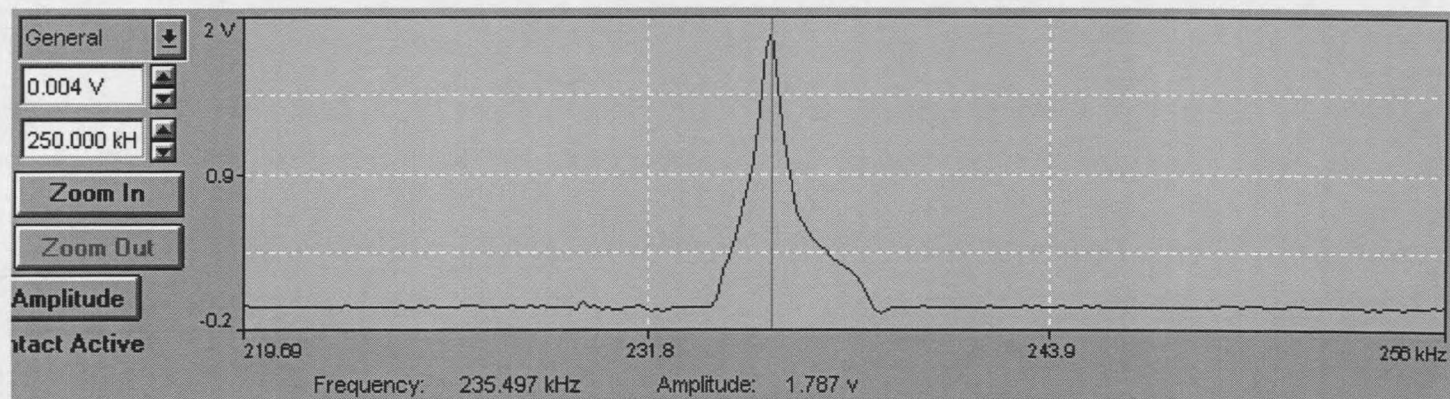


Figure 4.14. Selecting an operation frequency.

where Q is the quality factor of the cantilever, p is the piezoelectric gain of the modulating piezo (in distance/voltage), and A_e is the amplitude of the driving AC voltage.

Amplitude Detection Mode. One of the detection methods used in VCM is to detect a change in vibration amplitude, which is a result of the change in resonant frequency (Figure 2.4). The resonant frequency of the cantilever is

$$f_0 = \frac{1}{2\pi} \sqrt{\frac{k_e}{m_e}}, \quad (4.2)$$

where $k_e = k_0 + \partial F/\partial z$ is the effective spring constant, m_e is the effective mass, k_0 is the spring constant of the cantilever, and $\partial F/\partial z$ is the force gradient in the normal direction of the sample surface due to tip-sample interaction. When the tip-sample gap is sufficiently large, we have $\partial F/\partial z = 0$. Under normal conditions, such that only the van der Waals interaction exists, the tip-sample interaction appears to be more and more attractive when tip-sample gap is reduced, that is, $\partial F/\partial z < 0$. From equation (4.2), this force gradient reduces the effective spring constant of the cantilever, which reduces the resonant frequency of the cantilever.

In the VCM operation, the modulating piezo is applied with an AC voltage with constant driving frequency, f_D . Since f_D is originally set near the original resonant frequency ($f_D \cong f_0$) and $A(f_D)$ is the maximum amplitude, when the resonant frequency decreases, that is, $f_0' < f_D$, we have $A(f_D) < A(f_0)$. That is, a reduction of tip-sample gap causes a reduction of cantilever vibration amplitude. A stable feedback condition can be reached by setting the feedback set point to a certain amplitude value.

Phase Detection Mode. The phase detection mode is similar to the amplitude detection mode. In this mode, the photosensor signal, V_P , is multiplied by the driving signal, V_D , which modulates the cantilever. We have

$$V_D = \bar{V}_D \cos(\omega t) \quad (4.3)$$

and

$$V_P = \bar{V}_P \cos(\omega t + \varphi) \quad (4.4)$$

where φ is the phase shift between the two signals. The multiplication of these two signals gives

$$\begin{aligned} V_D V_P &= \bar{V}_D \bar{V}_P \cos(\omega t) \cos(\omega t + \varphi) \\ &= \bar{V}_D \bar{V}_P (\cos^2 \omega t \cos \varphi - \sin \omega t \cos \omega t \sin \varphi) \\ &= \frac{\bar{V}_D \bar{V}_P}{2} [\cos \varphi + \cos(2\omega t + \varphi)] \end{aligned} \quad (4.5)$$

When the double-frequency term is filtered and the vibration amplitude maintained constant, the DC term in equation (4.5) is only a function of the phase shift between the two signals. Since the phase shift is independent of vibration amplitude, this mode is especially useful for small-amplitude operation, in which the amplitude mode would have too low a signal-to-noise ratio for reliable measurements. In this mode, the signal goes through a phase-locked loop. Since only the signal of the same frequency as the driving signal is amplified, it suppresses the noise of other frequencies.

System Performance

To determine optimal operating conditions, the transfer function⁷¹ of the demodulator, which is the relation between the input signal amplitude and the output signal at various frequencies, is measured. Because of the intrinsic nonlinearity of electronic components, the linear region of the transfer function is limited. When the amplitude of the input signal is smaller than $V_{\max}$, the demodulated signal is proportional to the input signal amplitude. When the input amplitude is greater than $V_{\max}$, severe nonlinearity of the transfer function occurs. Since the nonlinear transfer function will cause significant imaging artifacts or unstable feedback, attenuation must be added to the input of the phase-locked loop when the input amplitude is too high.

The system operating frequency is from 50 kHz to 500 kHz. Figure 4.15 shows the transfer function in the amplitude mode, which was measured by using the internal frequency generator as the test source. The frequency and driving amplitude were all controlled by the software. For higher operation frequency, the demodulation output will be saturated for a smaller input signal. For example, when the working frequency is 180 kHz, the output will saturate for an input signal of 0.2 V. When the oscillation amplitude of the vibrating cantilever is greater than 20 nm, the signal from the photodetector is too strong. In this case, an extra attenuation circuit must be added to the input end of the modulation unit to keep the demodulator working in its linear range.

Figure 4.16 (a) is a typical amplitude signal of a resonance peak. For a normal driving amplitude there should be only one peak, which is the resonance peak of the cantilever. When the driving amplitude is large enough, other mechanical resonance of various components of the structure that supports and modulates the cantilever can be detected. Since these resonance modes have much smaller Q-factor, the resonance peaks are much smaller than the resonance of the cantilever, and can only be detected when a large amount of energy is transferred to the modulating piezo. If a large number of peaks are seen in the frequency spectrum, it is a good indication that the driving amplitude is too large. In this case, other parts of the scanner assembly are in resonance, which is not desirable. Figure 4.16 (b) shows the phase signal of all four quadrants.

Operation in Vacuum. Under vacuum, since the air damping no longer exists, the Q-factor of the cantilevers will increase dramatically, by about 100 to 1000 times. Although this could, in principle, increase sensitivity of the force sensor, the stability of the system will get worse. The proper detection method for VCM operation under vacuum should be amplitude detection. Since the Q-factor is very large in vacuum, the phase signal is very unstable. Given the inevitable large vibration amplitudes in vacuum, phase detection is also not necessary.

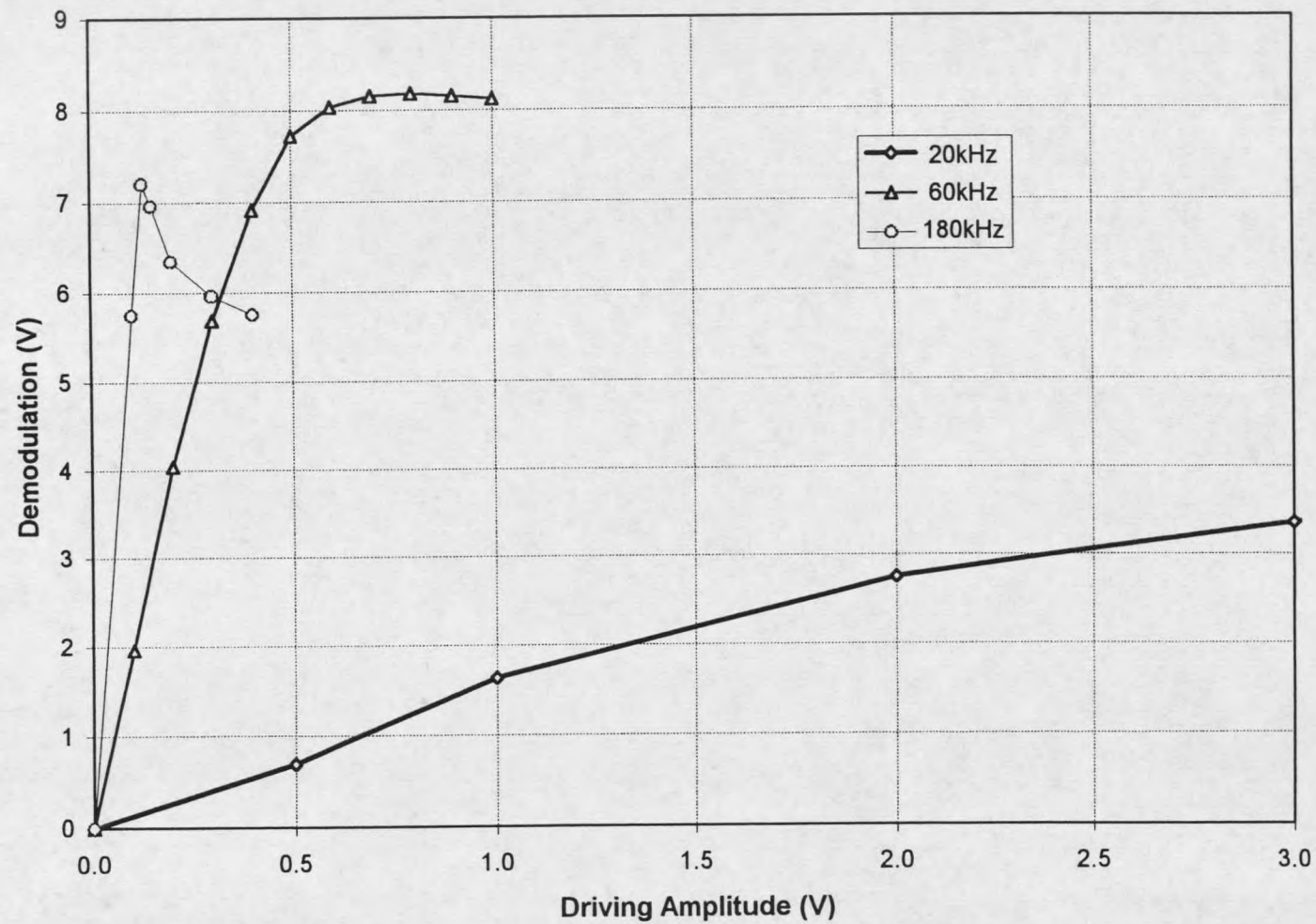


Figure 4.15. Transfer function of the control electronics in amplitude mode.

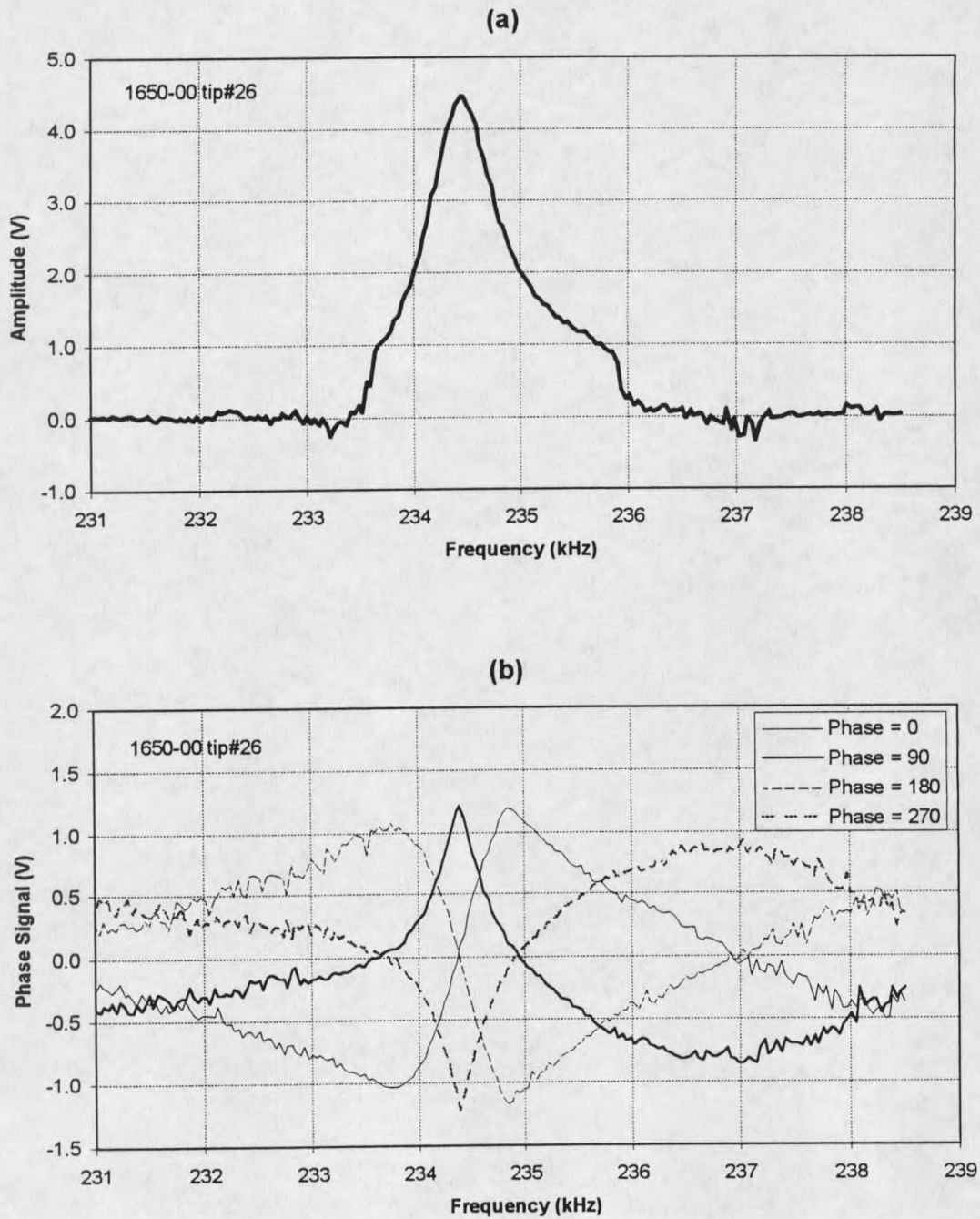


Figure 4.16. Frequency spectra of amplitude and phase signals.

Because the Q-factor is too high to have a stable peak, setting the driving frequency at the peak is not possible. A tiny fluctuation in the frequency generator can cause dramatic change in the vibration amplitude. To maintain stability, the drive frequency is set at a position greater than the resonant frequency $f_{dv} (>f_0)$, which is illustrated in Figure 4.17. Ideally, we should try to make the drive frequency, f_{dv} , satisfy the condition,

$$\left. \frac{dA_v(f)}{df} \right|_{f=f_{dv}} = \text{The maximum slope in air } \frac{dA_a(f)}{df}, \quad (4.6)$$

where $A_v(f)$ and $A_a(f)$ are the amplitude spectrum in vacuum and in air respectively. Because the operation at the maximum slope is stable in air, operating on the same slope in vacuum should give the same performance for the feedback electronics.

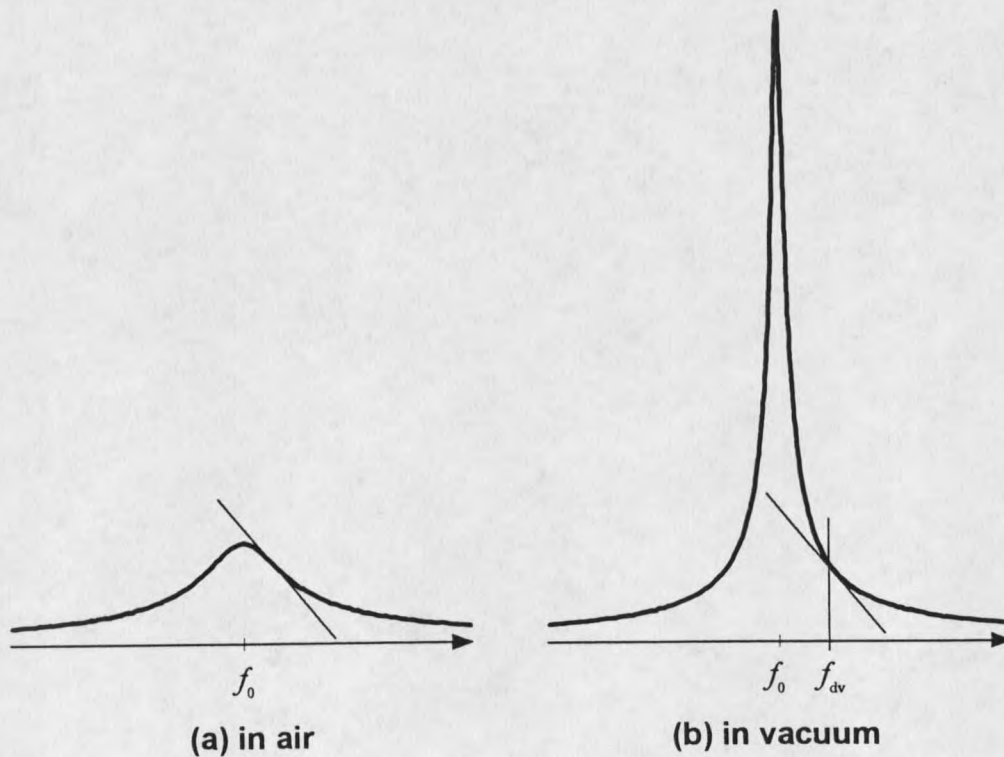


Figure 4.17. Selecting the drive frequency for vacuum operation.

Figure 4.18 shows a pair of VCM images acquired in air and in vacuum on an aluminum film sample. The sample was first imaged in air in the load-lock chamber. Then the load-lock chamber was evacuated and the image in vacuum was acquired 26 minutes later. The change of environment caused some drift in the AFM's field of view, which can be seen by location change of the same surface grain indicated by the white arrows.

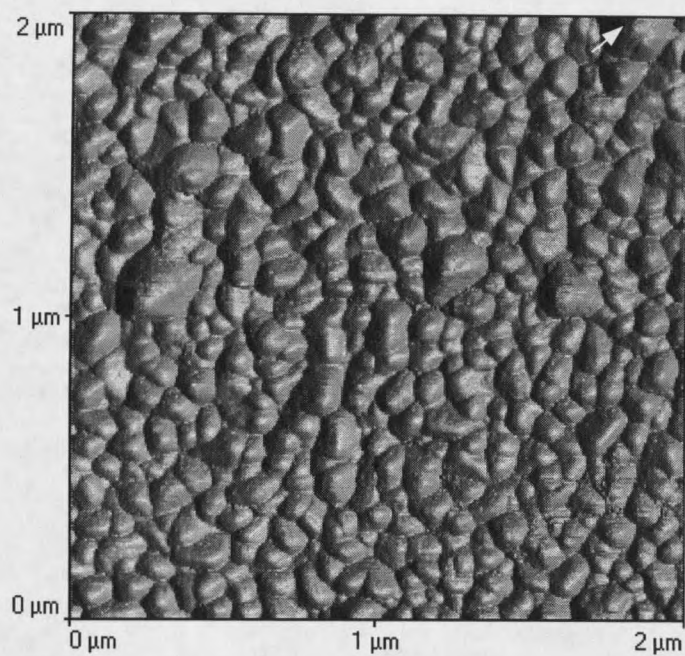
Double-Feedback-Loop Surface Potential Sensing System

For Kelvin probe force microscopy (KFM), a double-feedback-loop system was set up by two modulation units. Figure 4.19 shows the system diagram. Figure 4.20 shows the actual wiring of all the electronic units with the combined PPM/SEM system. The first modulation unit drives the Z piezo (or a separate modulating piezo) to generate the vibration of the cantilever. This feedback loop senses the resonant frequency change of the cantilever. It controls the Z piezo voltage to maintain a constant tip-sample gap. The Z piezo voltage generates the topographic image.

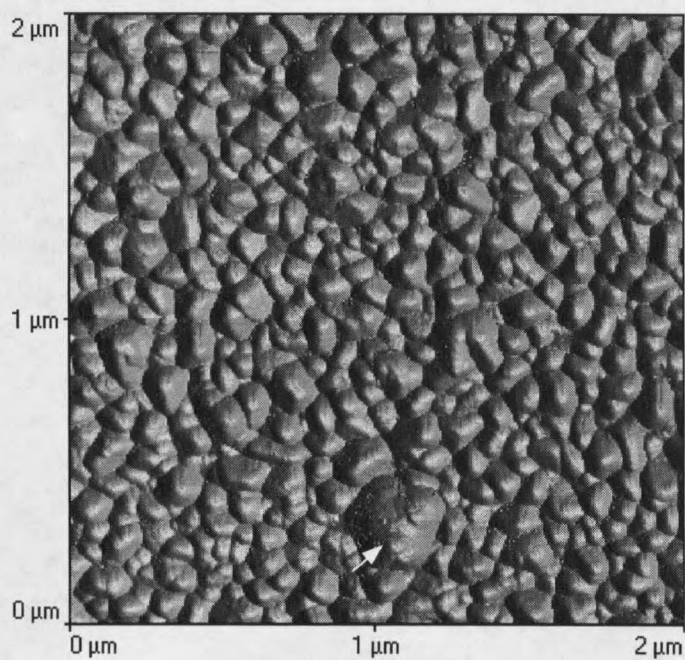
The second modulation unit applies the AC bias voltage onto the cantilever after the first feedback loop is established. It operates at a different frequency. It controls the DC bias on the cantilever to maintain the minimum phase shift. Thus, the electrostatic interaction between the tip and the sample is minimized. The DC bias voltage generates the surface electrical potential image. When scanning, these two feedback loops operate together. The topographic and potential images are generated simultaneously.

Potential measurement

To verify the proper operation of the potential imaging system, a method of calibrating the electrical potential was used. In this method, a metal coating sample was used to form a uniform surface potential, and a bias voltage was applied to this sample to create an adjustable electrical potential. During imaging, a sample bias voltage was manually changed every ten scan-lines by a step size of one volt.



(a)



(b)

Figure 4.18. Vibrating cantilever AFM images acquired in two environments: (a) in air; (b) in vacuum. The sample is an aluminum film on silicon substrate.

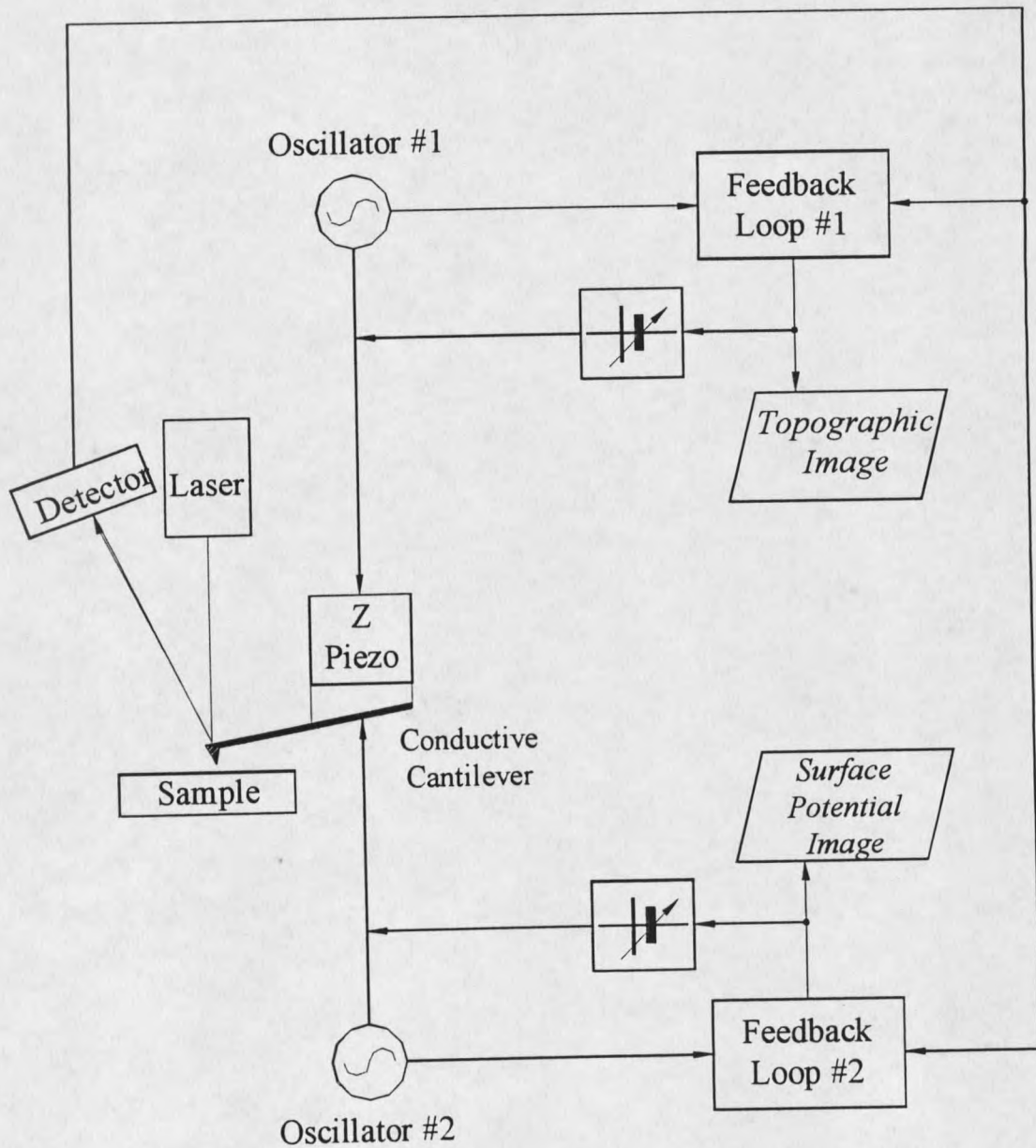


Figure 4.19. Diagram of the double-feedback loop surface potential sensing system.

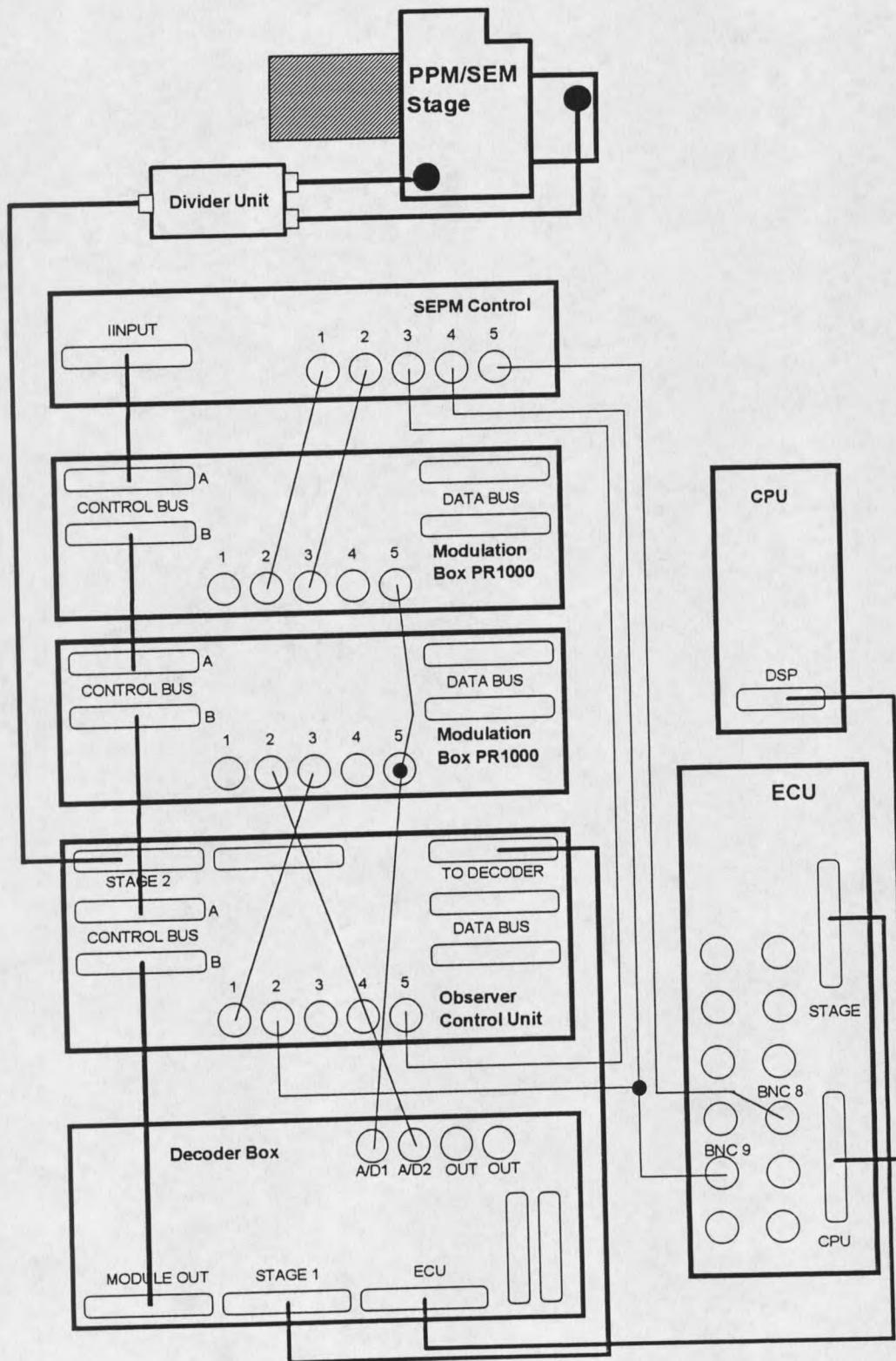


Figure 4.20. Wiring diagram of the surface potential imaging system.

Figure 4.21 shows the two simultaneously acquired images of surface topography and surface potential. The topography image shows a large number of small peaks about 15 nanometers in height, which are the results of the metal deposition process. The topographic image also reveals a piece of dust that is 100 nanometers in height. Although the surface potential was changed every ten lines, the topography showed no effect of such change. On the other hand, in the potential image, no obvious topographic features (even that outstanding piece of dust) are visible, which indicates that the cross-talk between the two feedback loops is minimal.

The maximum range of the DC bias used in the above measurement was ± 5 V. Since there is a small negative zero point offset, the potential signal overflows at -5 V sample bias. This caused the topographic feedback loop to not work properly. The instability in the topographic image can be seen on the scans indicated by the arrows on the sides of the images. From the potential image, the "surface roughness" measurement shows that the noise floor for the potential measurement is about 7 mV.

Spatial Resolution

To measure the spatial resolution of the potential image, an ideal sample should have a known microscopic potential structure at nanometer scale and without associated topography. However, such a sample is not available. Another method is to use a macroscopic structure having a sharp edge of potential change. By imaging a potential step, the spatial resolution can be estimated.

Figure 4.22 shows an example of macroscopic potential edge. The sample is a cross-section of a semiinsulating GaAs substrate with MBE grown overlayers. The interface is vertical across the image, where the left is the substrate and the right is the overlayer. A line profile is extracted from both the topographic and surface potential images. From the shape of the line profiles, we can see that the change of surface potential (in a step) is different from the change in topography (in a slope). This observation is important in the sense that the potential step is not due to the crosstalk between the two feedback loops, since the crosstalk would make both images look alike.

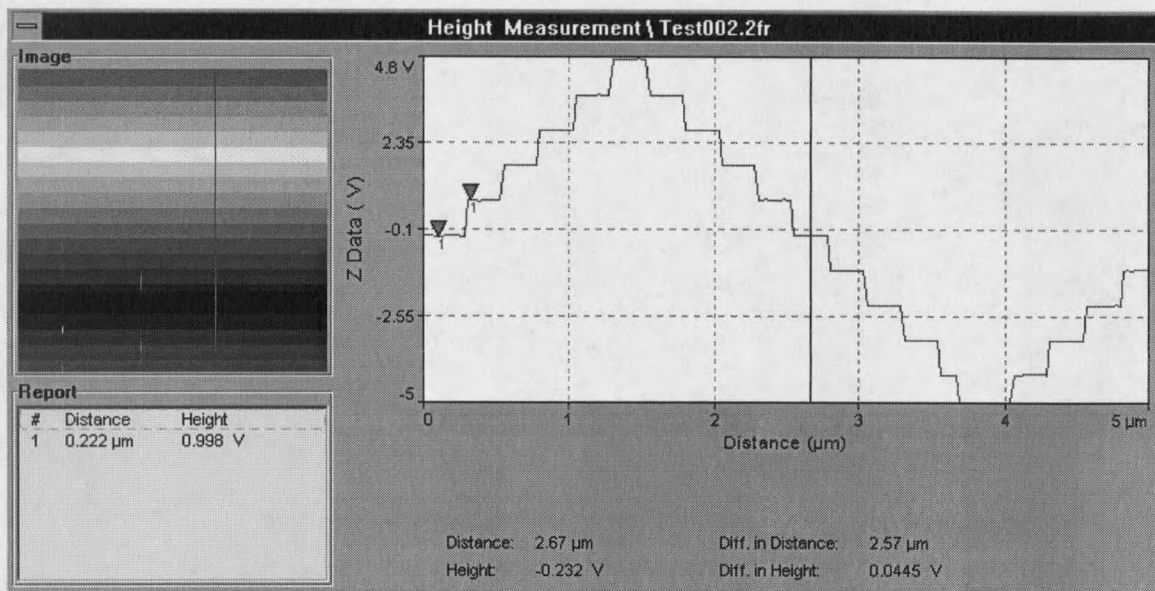
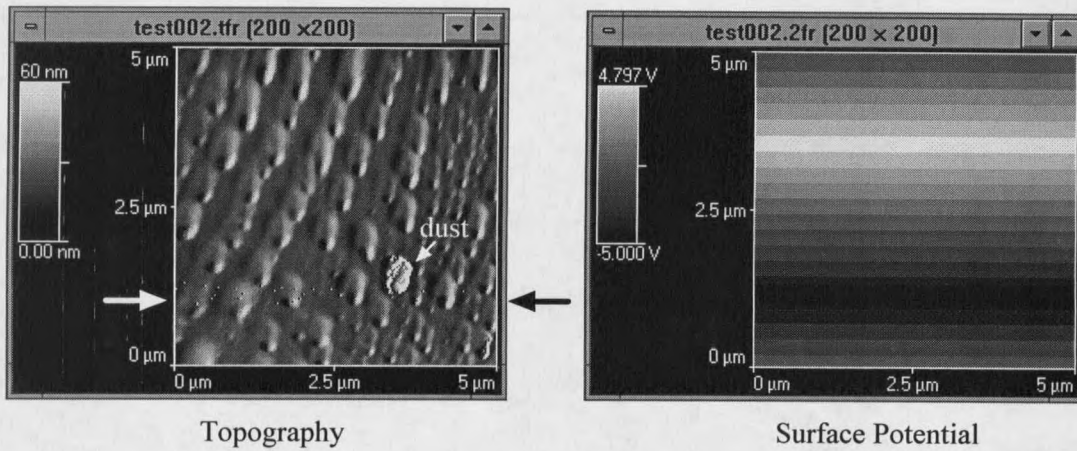
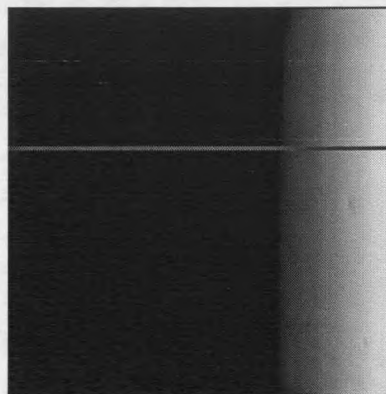
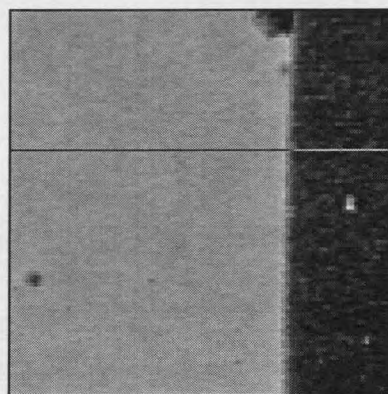
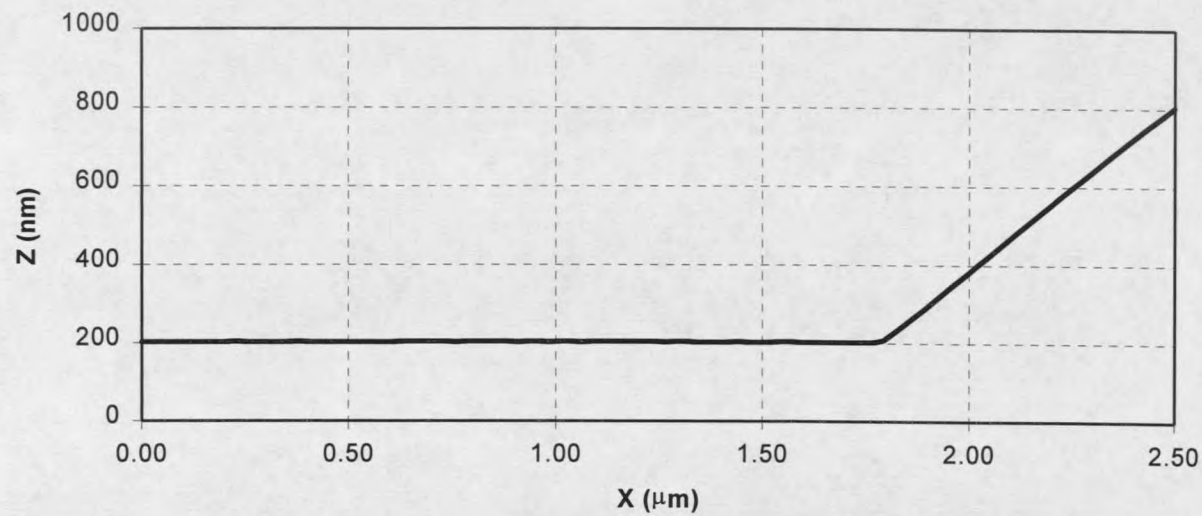


Figure 4.21. Testing the potential measurement.



(a) Topography



(b) Surface Potential

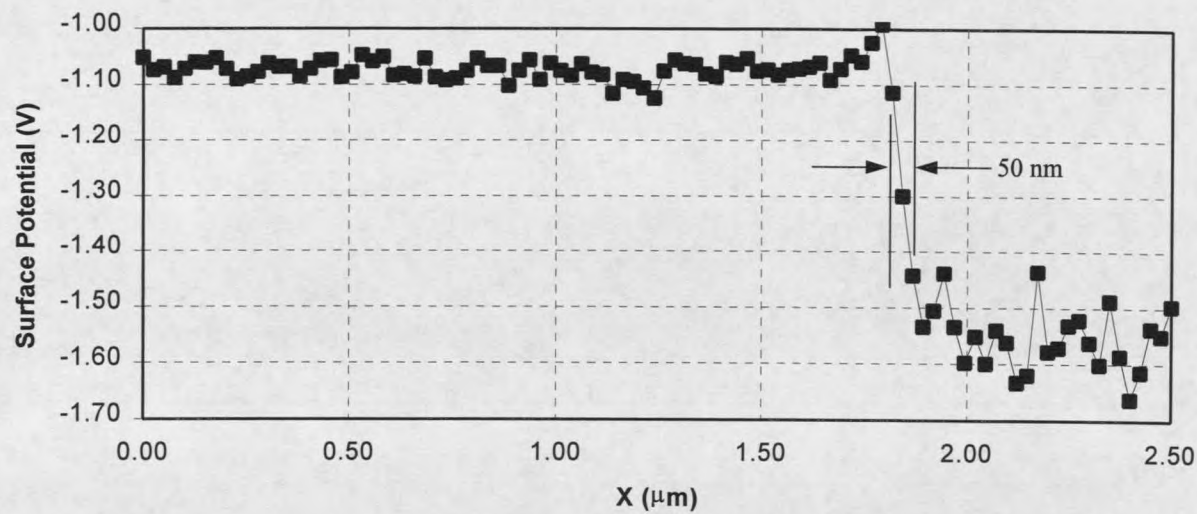


Figure 4.22. Spatial resolution of KFM

From the width of the step in Figure 4.22(b), we can estimate the spatial resolution to be about 50 nm. There are also a number of small features in the potential image that may be used to estimate the spatial resolution. However, since they have similar sharpness as the features show up in the topographic image, we can not rule out that they contain the effect of feedback loop crosstalk. Therefore, they are not used to estimate spatial resolution.

Operation Parameters

To minimize the crosstalk between the two feedback loops, the frequency difference between the two loops should be greater than the bandwidth of the PLL. However, when the driving frequency is too far away from the resonant frequency, the sensitivity will be reduced dramatically. Sometimes, when the cantilever Q-factor is too high, a smaller frequency difference has to be used. This could increase the crosstalk between the topographic signal and the potential signal.

AFM Probes

An AFM probe, consisting of a cantilever and a tip, is the central device for AFM. High quality images rely on high-quality probes. A number of these cantilever-based AFM probes are commercially available in different materials, spring constants and resonance frequencies for different applications. Three different types of probes were used in this research: silicon nitride, single-crystal silicon, and vapor-liquid-solid (VLS) silicon.

Silicon Nitride Probe

This is the first kind of cantilever produced with the IC technique. These cantilevers are usually "V" shaped. The length of the cantilevers range from 100 to 200 μm . The spring constant is usually less than 1 N/m, and the resonant frequency is usually less than 20 kHz. The probes are of pyramid shape. Since the cantilever is soft and the tip is tough, this kind of probe is mostly used for contact mode operation.

The silicon nitride probes initially made from the integrated circuit wafer process are not very sharp. Their radii are usually greater than 50 nm. A number of tip sharpening techniques were developed to sharpen the tip, and increase the aspect ratio, at the same time. One is to grow a carbon tip by a focused electron beam at the end of the silicon nitride tip.⁷² A focused ion beam (FIB) is also used to mill down the side of the pyramid to leave a high aspect ratio tip (Figure 4.23(a)).

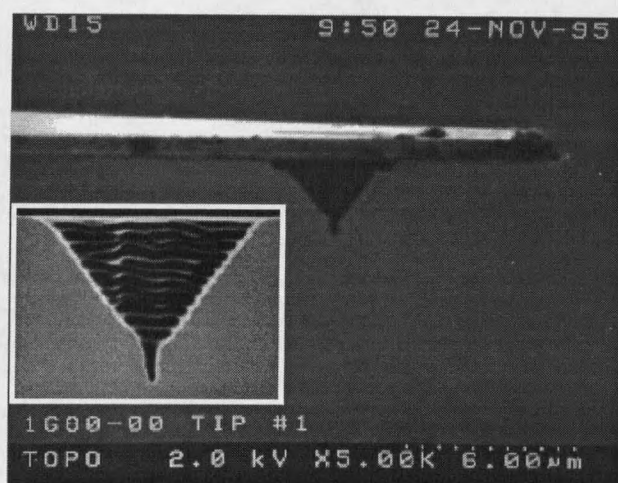
Silicon Probe

This kind of probe is etched out of a silicon single crystal. The tips are, in general, sharper than the silicon nitride tip (see Figure 4.23(b)) with radii of less than 5 nm for most of them. The cantilever is rectangular in shape with a length ranging from 100 μm to 450 μm . The resonant frequency of the cantilever can be up to about 1 MHz.

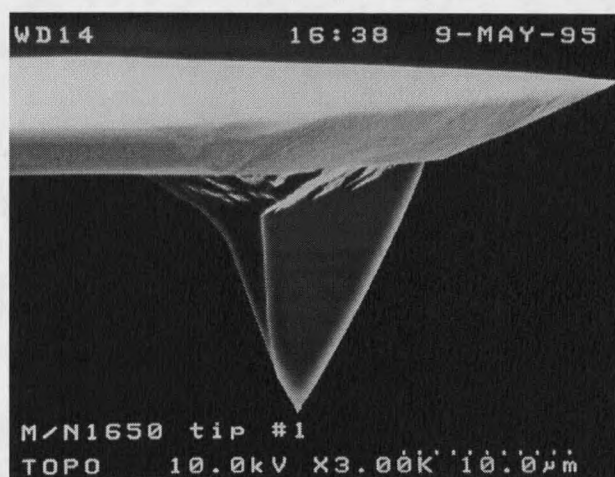
The probes with higher resonant frequencies are used for vibrating cantilever mode (VCM) operation. Since the silicon tip is not very robust, contacting the surfaces can result in serious tip damage. In VCM with appropriate operating parameters, the tip can be controlled so that it never contacts the surface.

VLS Silicon Probe

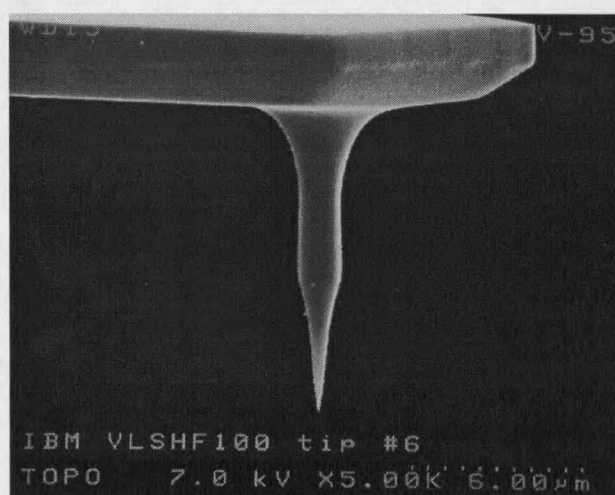
The tip is grown near the end of the rectangular cantilever by the vapor-liquid-solid (VLS) method.^{73, 74} The cantilever for the VLS silicon probe is also a silicon single crystal. The advantage of these tips is their high aspect ratio and small cone angle (see Figure 4.23(c)), which are important characteristics in imaging rough samples or deep structures. The aspect ratio is usually greater than 10:1 and the cone angle is usually smaller than 10° . Because the tip is long, it also allows tip viewing from the 45° which is important for operating in the AFM/SEM combined system. With the capability of 45-degree viewing, placing the tip on top of the desired feature is straightforward.



(a)



(b)



(c)

Figure 4.23. The AFM probes used in this dissertation. (a) Silicon nitride with FIB sharpening; (b) Single crystal silicon probe; (c) Silicon tip grown with the VLS technique.

Summary

The experimental apparatus presented here is mostly for studying nanometer-scale surface interaction. The methods of vibrating cantilever AFM and double-feedback-loop surface potential measurement are previously published. The totally new experimental apparatus, built in this thesis work, is the combined PPM/SEM instrument. It is the first optical-deflection AFM built into a SEM. Besides that, it is also a full-function PPM, which consists of two levels of XYZ translation for tip and sample positioning, and is capable of operating both STM and AFM, and their various modes. A technique was developed for operating VCM in vacuum, which overcomes the instability problem caused by the extremely high Q-factor of the cantilever. A method for calibrating the potential measurement for KFM using a controllable sample bias was demonstrated.

CHAPTER 5

STUDIES OF AFM LOADING CURVES

Introduction

This is the first chapter that deals with the experimental results on nanometer scale tip-sample interaction studies. The studies of this chapter focus on the force dependence on the tip-sample gap. The force-sensing tip of an atomic force microscope (AFM) is mounted on the free end of a cantilever, which measures the vertical component of the tip-sample interaction force. To study the tip-sample interaction, the magnitude of the force can be measured while changing the tip-sample gap. This measurement is called a loading curve, or a force-distance curve. All the experiments in this chapter were performed with this simple method to probe the tip-sample interaction.

An objective of this chapter is to understand the surface contamination layer that is always presence on a sample surface in ambient air. Since the surface contamination layer is liquid-like, it creates a capillary force that plays an important role in the tip-sample interaction. Studying the loading curves can provide information about the properties of this contamination layer. A method used here to study the surface contamination is to compare the loading curves measured in air and in vacuum. Since the liquid-like contamination layer is removed under vacuum, the comparison gives us information on the contamination under ambient air.

Studying the surface contamination is critical for understanding the imaging mechanism of PPM in ambient air. In order for a proximal probe microscope (PPM) to work, the tip must be kept

at close proximity to the sample surface during imaging, where the contamination on the tip and the sample will interact with each other.

In the following *Experiments* section, we will first explain the detail of measuring nanometer scale force and loading curves with an AFM. Next, we show the method of calibrating an experimental loading curve for theoretical comparison. Then we discuss the meaning of a loading curve. Finally, we describe the method of comparative measurement in air and in vacuum.

In the *Results and Analysis* section, we first discuss the reproducibility issue and conclude the factors that are responsible to the reproducibility problem. Then we show the result of comparing of cantilevers of different spring constants to demonstrate the condition of JTC, as we have discussed in Chapter 3. Finally, we describe the fitting of an experimental approach curve with the theoretical model of capillary force (described in Chapter 3), to obtain the thickness of the contamination layer and the magnitude of the capillary force.

Experiments

Measuring Force with a Micro Cantilever

The cantilever-based optical deflection force sensor uses the bending movement of a micro cantilever to detect tip-sample interaction force. Figure 5.1 shows the basic structure of this force sensor. Distance $\bar{Z}$ is the sample location, relative to the fixed end of the cantilever, whose change is equal to the piezo movement. $\bar{D}$ is the amount of vertical displacement of the cantilever free end due to tip-sample interaction, and $\bar{G}$ is the actual tip-sample gap used in the theoretical model. Therefore, we have

$$\bar{G} = \bar{Z} - \bar{D}. \quad (5.1)$$

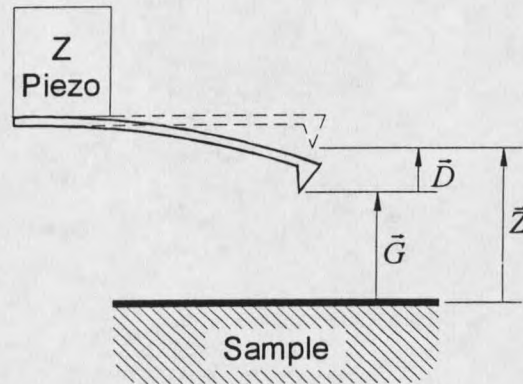


Figure 5.1. Diagram of the cantilever bending.

The displacement of the free end of the cantilever is proportional to the force applied to it.⁷⁵ The optical deflection sensor detects the change of the angle at the free end of the cantilever. Since the bending angle is small, the angle can be considered to be proportional to the displacement of the free end of the cantilever. Therefore, we can consider that the force sensor is measuring the vertical displacement, $\vec{D}$. Since the forces between the tip and the sample are mostly attractive before the tip and sample contact, the cantilever usually bends towards the sample.

Figure 5.2 shows the mechanism of force measurement by the deflection of the cantilever. The change in tip-sample interaction causes the free end of the cantilever to move. A laser beam is directed to the back of the free end of the cantilever by the secondary mirror, which is then deflected by the movable cantilever back up to the secondary mirror, the primary mirror, and finally the photosensor. The photosensor includes two segments, A and B , that can generate electric voltages depending on the number of photons received. Since the laser beam is focused at the cantilever, and the beam spot gets defocused at the photosensor so that it covers both segments of the photosensor, the segments A and B thus generate photovoltage, V_A and V_B , respectively. The difference of the photovoltage, $V_A - V_B$, gives a measurement of the location of the center of the laser spot, which in turn gives a measurement of cantilever displacement.

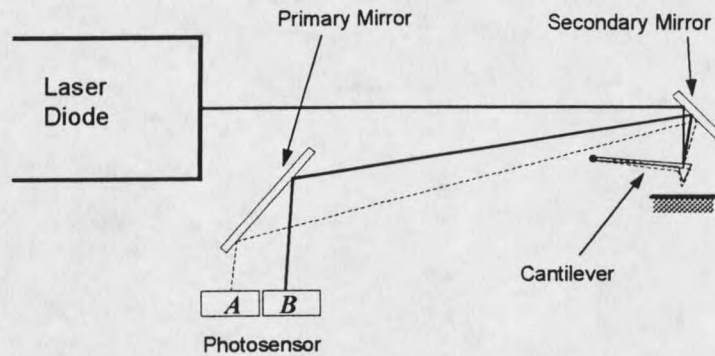


Figure 5.2. Mechanism of force detection with the optical deflection force sensing system.

Measuring a Loading Curve

When a control voltage is applied to the Z piezo to change the tip-sample gap, the variation in force can be measured by changes in the difference of the photovoltage, $V_A - V_B$, which produces a loading curve. The experimental procedure for measuring a loading curve is the following:

- 1) The tip is brought into contact with the sample surface with the feedback loop on.
- 2) While maintaining the Z piezo voltage, the feedback loop is turned off.
- 3) The Z piezo moves the tip away from the sample. At this point, the computer is prepared to record the force signal.
- 4) The Z piezo first pushes the tip toward the sample a certain distance, then it pulls the tip back to the starting point to complete a full cycle of movement. During the tip movement, the change in $V_A - V_B$ is recorded.

Figure 5.3 shows a typical loading curve. The vertical axis represents the force detected by the cantilever. The zero force is the force value when the tip-sample gap is sufficiently large. The horizontal axis represents the position of the Z piezo that controls the tip-sample gap. The zero point, which is believed to be the ideal point where the tip starts to contact the sample surface, is determined by the point where the force changes from negative (attractive) to positive (repulsive) in the approach curve.¹⁰

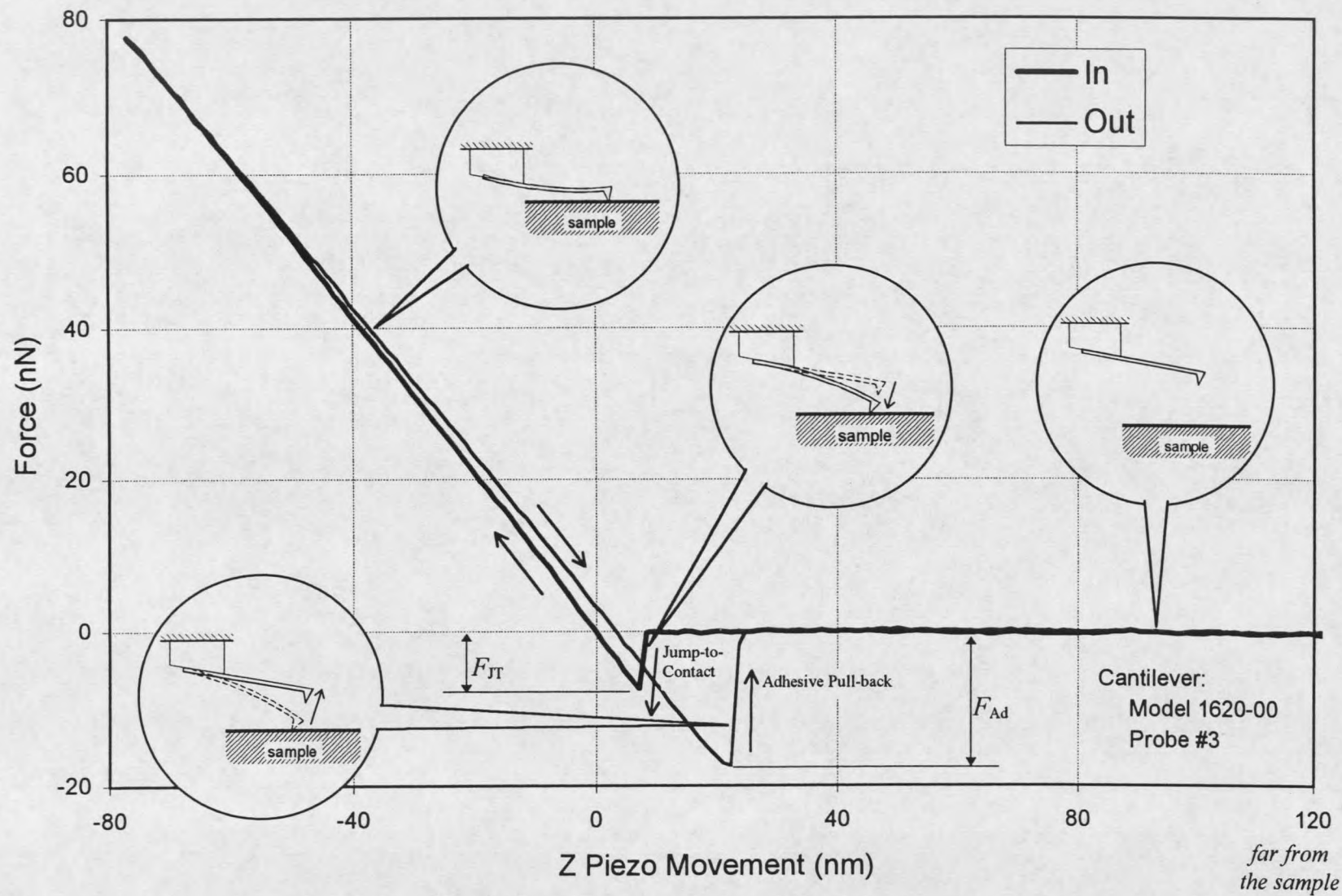


Figure 5.3. An experimental loading curve.

Calibrating a Loading Curve

The vertical axis of a loading curve represents the force experienced by the cantilever. To calibrate the loading curve, we must first calibrate this force. The relation between the cantilever displacement, ΔD , and the force applied to its free end, ΔF , needs to be determined as

$$\Delta F = k\Delta D, \quad (5.2)$$

where k is the spring constant of the cantilever.

The electrical voltage signal from the AFM force sensor that is proportional to the force needs to be converted to a number in the unit of newton. This can be done in two steps. First, we need to calibrate the change in photosensor output signal, ΔV , with respect to the actual cantilever displacement, ΔD . That is

$$\Delta D = g\Delta V, \quad (5.3)$$

where g is the gain factor of the optical deflection force sensor.

The first coefficient, g , can be determined experimentally by bringing the tip into contact with the sample. To do this, a hard surface sample is used, and we assume that the sample surface is infinitely rigid. With this assumption, the repulsive force is sufficiently large when the tip contacts the sample, so that the tip-sample gap $G = 0$. Thus from equation (5.3) we have $\Delta Z = g\Delta V$. Since ΔZ , which is the Z piezo displacement, and ΔV are both measurable, the coefficient g can be calculated.

The rigid surface assumption can be easily satisfied under most circumstances, because the actual indentation of an AFM tip to a "hard" sample, such as silicon, is on the order of a few nanometers or less. Compared to thousands of nanometers in cantilever movement, it is reasonable to assume that the tip-sample gap $G = 0$, when the repulsive force is great enough. Actually, the high repulsive region is very linear in most of the experimental loading curves, which indicates that

G is truly negligible.

The second coefficient to be determined is the spring constant k of the cantilever. The silicon cantilevers used for force measurement are rectangular. Their geometric dimensions can be measured with SEM, and the spring constant can be calculated by⁷⁶

$$k = 3EI/l^3, \quad (5.4)$$

where E is Young's modulus, l is the length of the cantilever, $I = wt^3/12$, w is the width, and t is the thickness. Since we have calibrated the displacement measured, and we now have the spring constant, the force can be determined. Combining equations (5.3)-(5.4), the tip-sample interaction force can be calibrated.

An example of loading curve calibration is shown in Figure 5.4. In an unprocessed experimental loading curve (Figure 5.4a), the horizontal axis represents the Z piezo position, Z , which can be directly measured. Therefore, this calibration choice is simple. However, in theoretical analysis, the tip-sample gap, G , is commonly used as the independent variable of the force, which would prefer the horizontal axis in the loading curve to be plotted as the tip-sample gap. From Figure 5.1, we can see that the Z piezo position is the same as the fixed end of the cantilever, and the tip-sample gap is measured at the free end of the cantilever. Thus the relation between the Z piezo position and tip-sample gap is governed by the cantilever displacement, D . Since the cantilever is not a rigid body, it will flex depending on the tip-sample interaction force, which causes the change in Z piezo position to be different from the change in tip-sample gap. Therefore, this calibration choice is not straight forward. According to equation (5.1), the original loading curve can be processed so that the horizontal axis is G . Since this calibration removes the cantilever deflection effect from a loading curve, we call this loading curve to be cantilever-deflection corrected (Figure 5.4b).

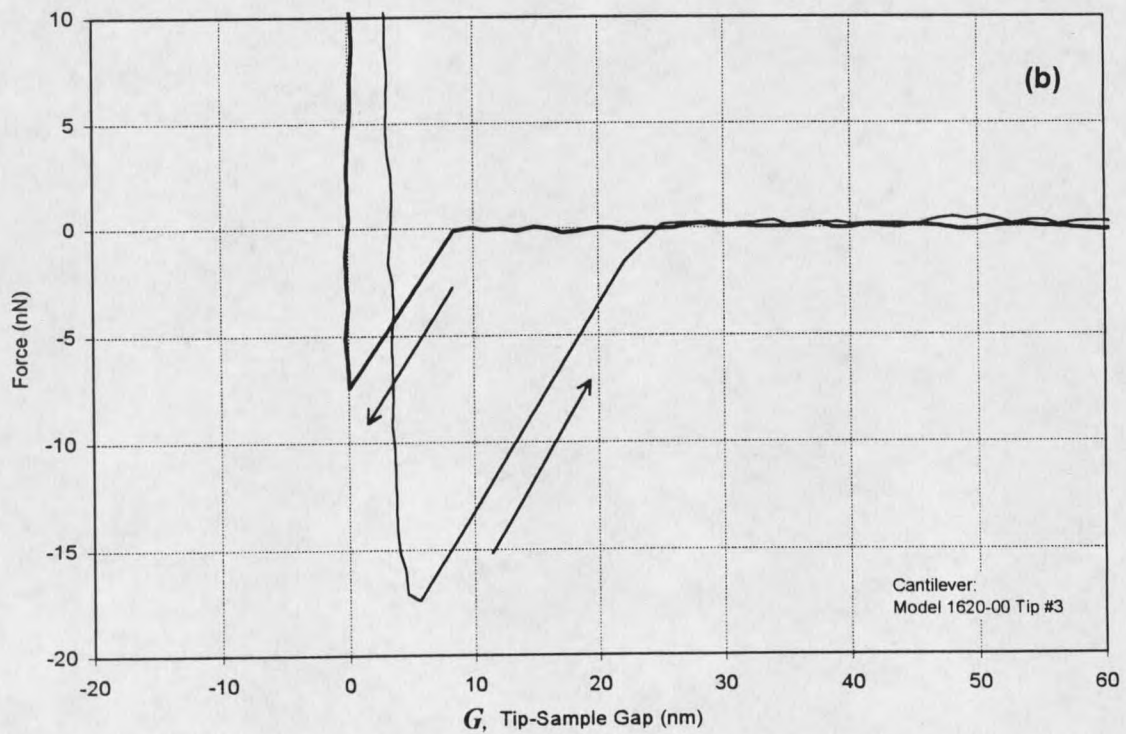
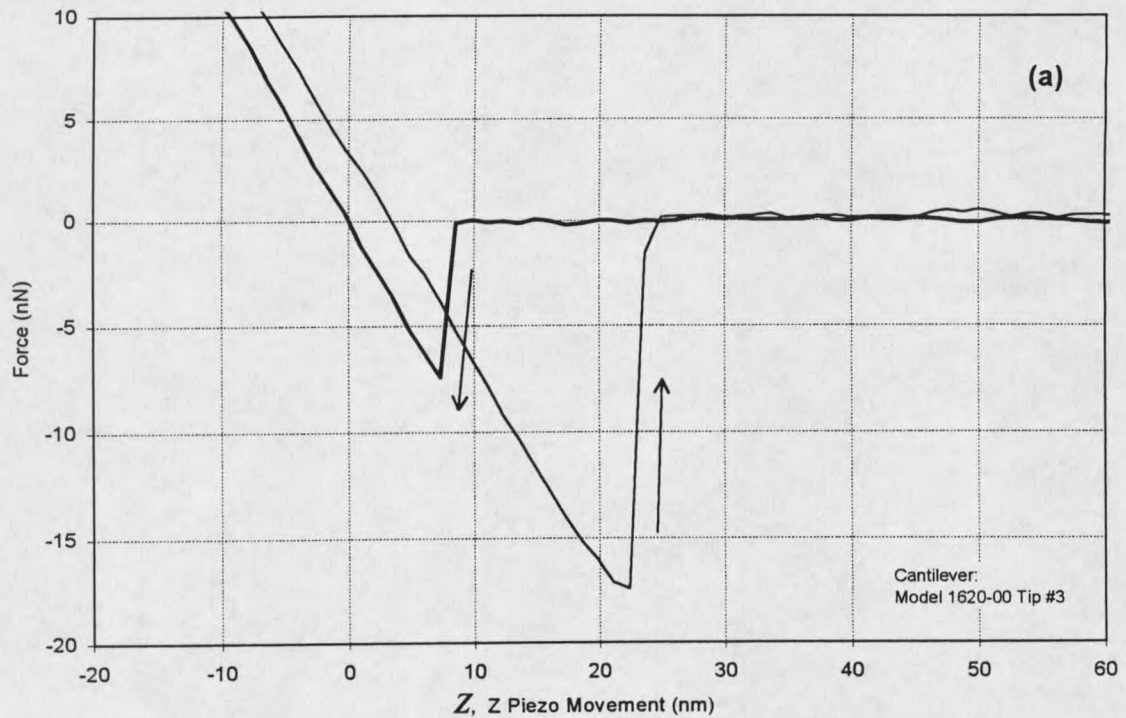


Figure 5.4. An example of loading-curve calibration using a loading curve measured in air: (a) As measured. (b) Cantilever-deflection corrected from the measured curve in (a).

Interpreting a Loading Curve

The two most common characteristics of loading curves observed in most of the measurements, shown in Figure 5.3, are the jump-to-contact (JTC) and the adhesive pull back.⁷ These are the abrupt movements of the free end of the cantilever, in which the free end jumps to a new position, even if the other end, which is fixed to the Z piezo, stays at the same position. They can be quantified by two parameters, F_{JTC} and F_{Adh} (see Figure 5.3). F_{JTC} is the amount of abrupt force change in the approaching curve, while F_{Adh} is the amount of abrupt force change in the pull-back curve, which is also called *adhesion*.

The origin of these abrupt cantilever movements can be explained by comparing the spring force of the cantilever with the tip-sample interaction force. When the cantilever moves towards the sample, the cantilever deflects so that the spring force balances the tip-sample interaction force, that is,

$$|F_{TS}(x)| = |F_{SP}(x)| = k(x-x_0), \quad (5.5)$$

where $F_{TS}(x)$ is the tip-sample interaction force, $F_{SP}(x)$ is the spring force due to the cantilever bending, k is the spring constant of the cantilever, x is the tip position, and x_0 is the tip position when the spring force is zero. From Figure 5.5 we can see that when the tip-sample gap gets smaller, the slope of the $F_{TS}(x)$ curve increases. When the tip moves from point D to point A, the rate of increase of $|F_{TS}|$ is small enough so that for whatever the change in $|F_{TS}|$, the spring can deform to create a large enough $|F_{SP}|$ to balance the system. That is, the system can stay in balance whenever the derivative,

$$dF_{TS}(x)/dx < k. \quad (5.6)$$

However, when the tip moves beyond the point A, the increase of $|F_{TS}|$ is so large that the increase of $|F_{SP}|$ cannot catch up. That is, if $dF_{TS}(x)/dx > k$, the balance will be broken. At point A, any increase in cantilever deformation will result in greater $|F_{TS}|$, which in turn deforms the cantilever further. At that moment, the cantilever's free end jumps to a new position where $dF_{TS}(x)/dx < k$, so

that the cantilever can establish a new balance. When the tip moves to point C from point B, the reverse mechanism occurs, so that the tip will jump to point D to stay in balance.

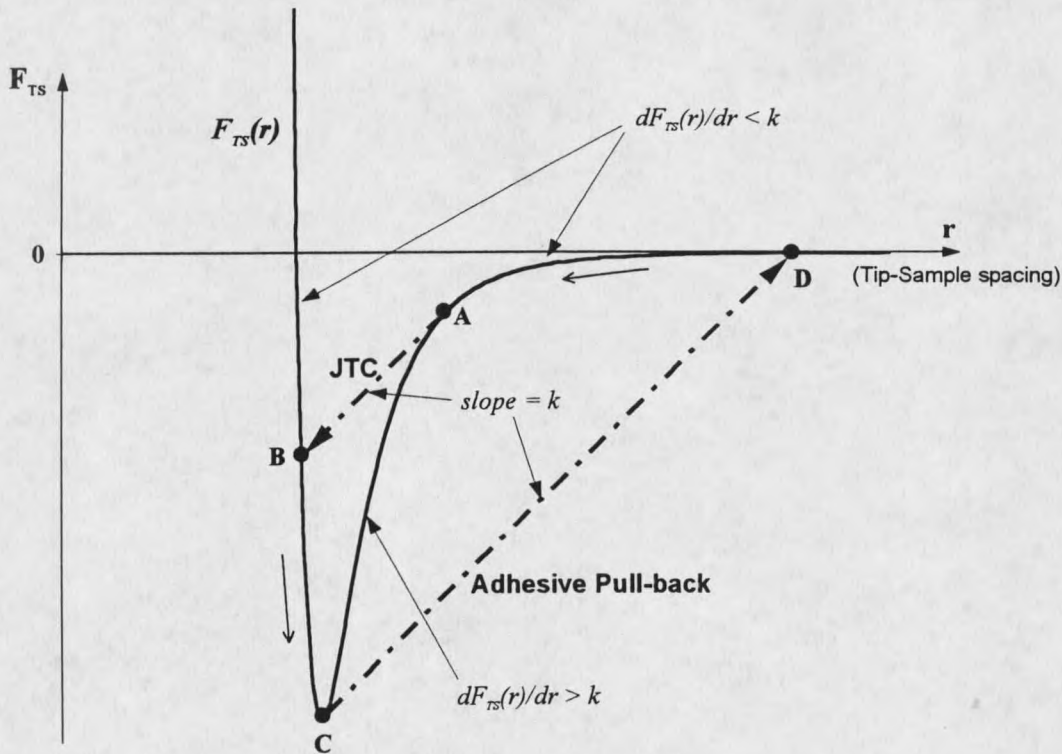


Figure 5.5. Jump-to-contact and adhesive pullback.

In the experimental loading curves, it can be seen that the approach curve and the pullback curve cross one another. This is caused by the relaxation effect of the Z piezo ceramic. When the control voltage applied to the Z piezo changes from forward to backward, the forward movement of the Z piezo is not stopped instantly. This artifact in the experimental loading curve is not easily removed.⁷⁷

Comparing Loading Curves in Air and in Vacuum

Currently, most AFMs are operated in air. When operating in air, the tip-sample interaction is

strongly governed by the contamination layer on the surface. The instability of this contamination layer may be a source of fluctuation in the loading curve measurement. When the sample is brought into vacuum, most or all of this contamination layer is removed. In order to understand the influence of this contamination layer, comparative studies of loading curves in air and in a medium vacuum ($<1 \times 10^{-3}$ Torr) were performed.

The experiments were done in the load-lock chamber of the combined AFM/SEM system. The load-lock chamber can be evacuated or vented to air independently, so the AFM unit can be operated in air or in vacuum without changing the tip-sample relation. The AFM loading curves were measured in both environments on the same spot so as to eliminate the influence of surface topography variation and surface material.

Results and Analysis

Reproducibility of the Measurements

Major factors that contribute to the result of a loading curve measurement are: the tip, the sample, the contact location on the sample, and the environment. When a set of loading curves are measured consecutively with the same tip, the same sample, at the same contact point, and under the same environment, the results are generally repeatable. This reproducibility can be revealed by the measurement of F_{TC} and F_{Ahd} . Figure 5.6 shows two sets of consecutive measurements on an aluminum mirror sample measured in air and in vacuum. The spikes in the vacuum measurements are caused by lack of air dampening when the tip is being pulled away from the sample.

Any change in the above conditions can dramatically change the result of the loading curve measurement, creating apparently poor reproducibility. To study the effects of various factors, systematic loading curves were measured. Figure 5.7 shows a set of results measured with the same tip on three different samples. The results on two different locations and under two different environments are shown.

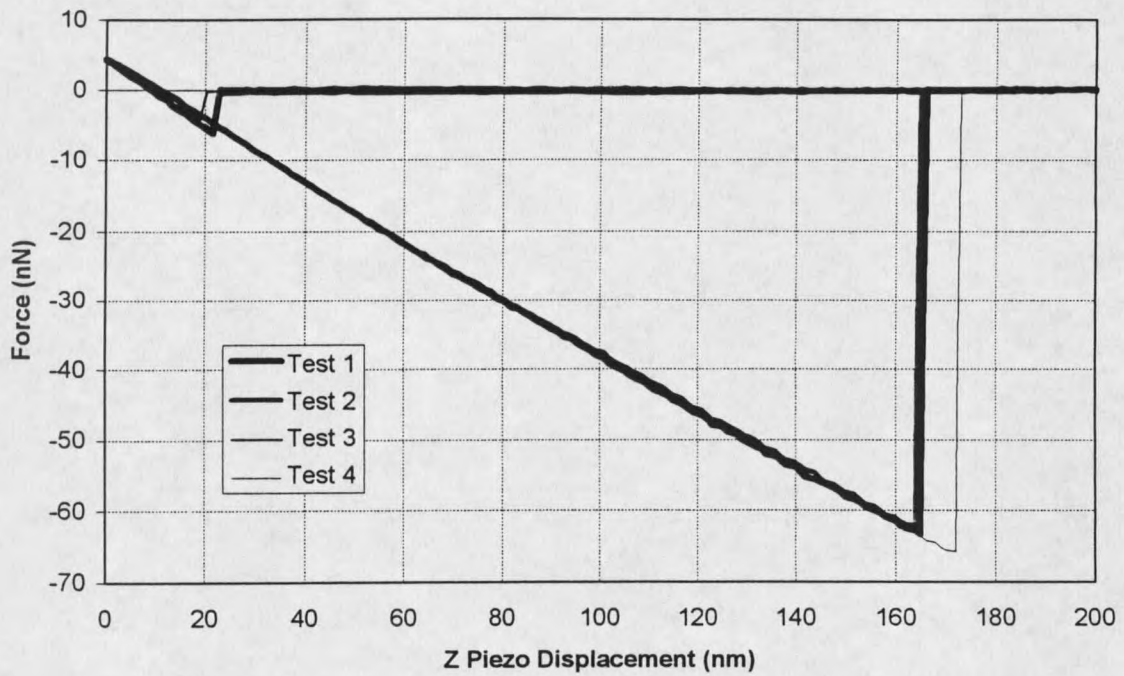
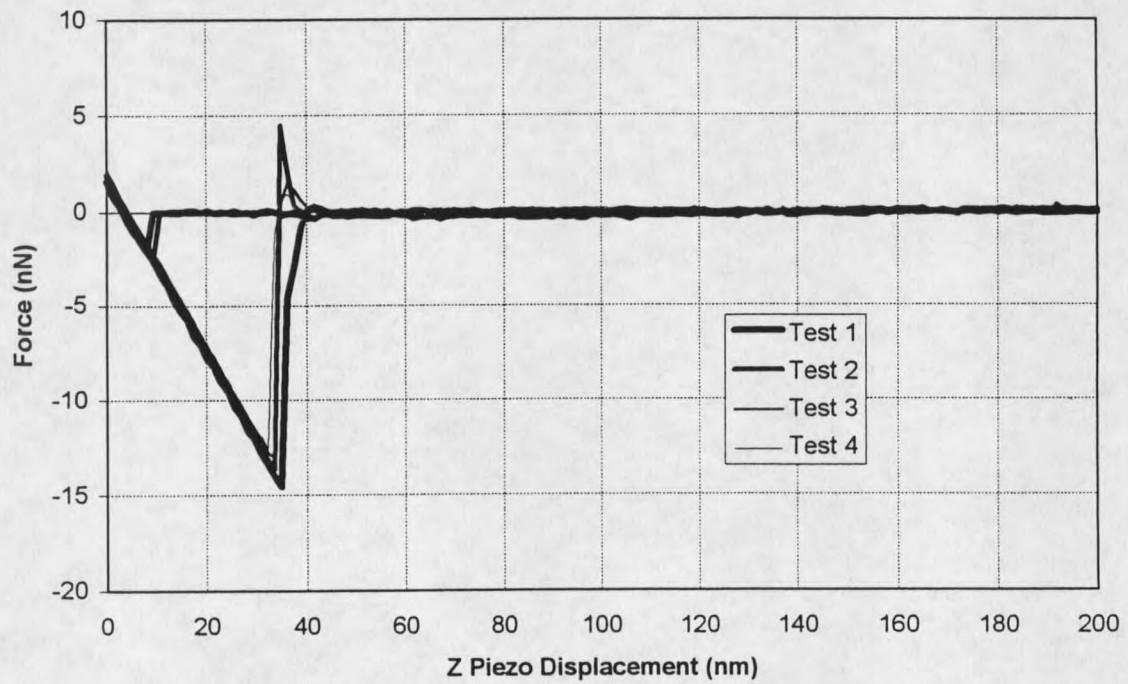
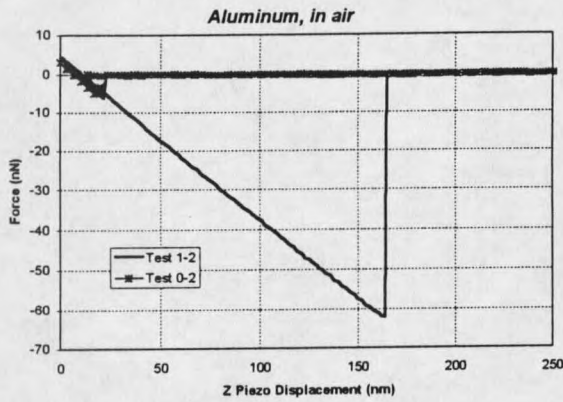
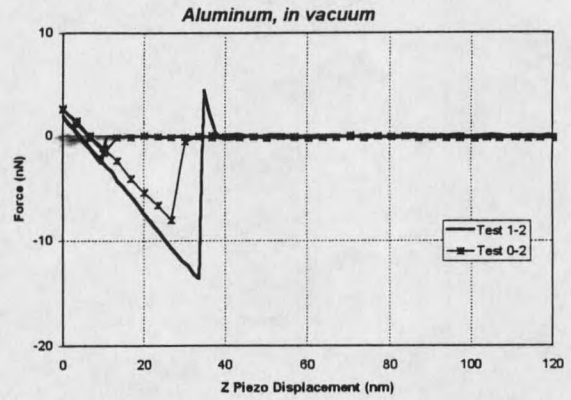
In Air*In Vacuum*

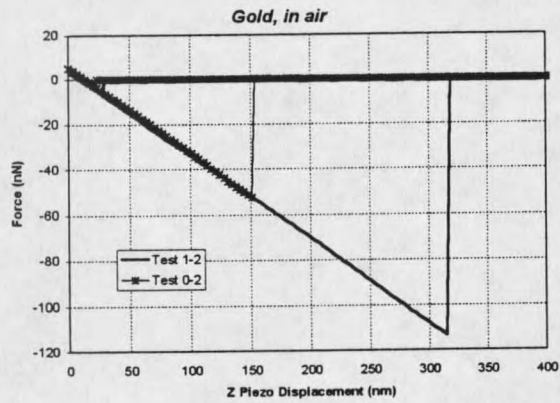
Figure 5.6. Repeatable loading curves measured consecutively.



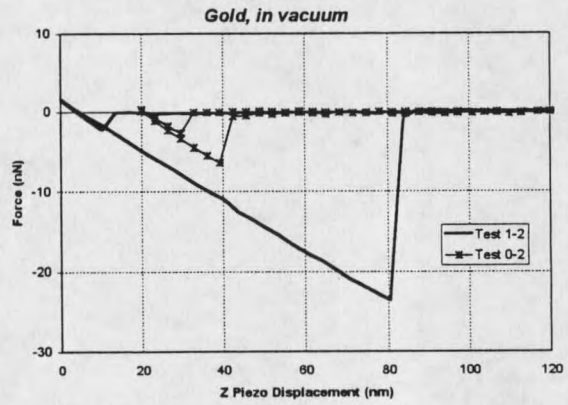
(a)



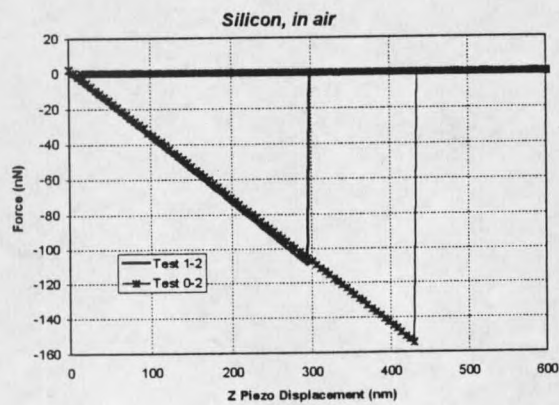
(b)



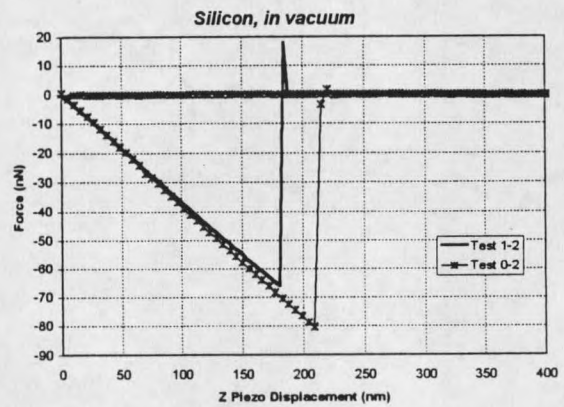
(c)



(d)



(e)



(f)

Figure 5.7. Loading curves measured on different samples.

The major factors that affect the reproducibility of a loading curve measurement are:

- 1) *Effect of changes of the tip.* Because the loading curve measurement involves tip-sample contact, which can apply large pressure to the tip and the sample surface in a nanometer scale area, the shape of the tip and surface condition can change after each measurement. As measurement continues, the tip will be changed even further.
- 2) *Effect of surface topography.* Since tip-sample interaction depends on the local slope of the topography, different surface locations will have different contact conditions.
- 3) *Effect of surface contamination.* Surface contamination applies a capillary force on the tip when it is in contact. Since water is a important component in the surface contamination layer,⁷⁸ changes in humidity can also affect the results of loading curve measurements.
- 4) *Effect of surface material.* The type of surface material mostly affects adhesion when the tip is pulled away from the sample. After making physical contact, the sample material strongly influences the tip-sample interaction. Therefore, the measurement of adhesion can be used to distinguish materials.⁷⁹

Consistency in tip-sample interaction is difficult to achieve in the contact condition, which causes modification of the tip and the sample surfaces. With the combined AFM/SEM system, tip damage was observed after a contact measurement. A sharp AFM tip is seriously blunted during the first set of loading curve measurements, even though the loading force did not exceed 10 nN. When the tip radius is significantly smaller than the surface features, the surface roughness will cause non-repeatable results on different areas of the surface.

To obtain consistent reproducible loading curve results, a spherical tip with a large tip radius (12 μm) had been used by other researchers.⁸⁰ The advantage is that it will not be damaged as easily as a sharper tip, and the influence of the surface topography will be smaller. However, the surface interaction is not in the nanometer scale due to the large contact area.

From the discrepancies in the adhesion measurements on three different surfaces shown in

Figure 5.7, we can see that the reproducibility of the adhesion measurement is better when the loading curves were measured under vacuum environment ($\sim 5 \times 10^{-4}$ Torr). Although not obvious when comparing Figure 5.7(c) and Figure 5.7(d), from the absolute changes in the magnitude of the forces, we can see that the reproducibility in vacuum is better for the gold sample. Since the liquid-like contamination layers was removed in vacuum, the improvement in the reproducibility indicates that the surface contamination layer is an important factor causing poor reproducibility of the loading curve measurement. The fluctuation in vacuum is only due to the change in contact locations.

Loading Curves for Cantilevers of Different Spring Constants

In Chapter 3 it is shown that the smallest resolvable size is proportional to the tip-sample gap. To get a higher-resolution image, the tip needs to be closer to the surface. However, a major problem in vibrating cantilever mode (VCM) imaging in air is the JTC that occurs when the tip is brought close to the surface for optimizing lateral resolution. When JTC occurs, the tip is captured by the sample surface and cantilever vibration stops, which disables VCM operation. Considering that JTC is due to the capillary attraction of the contamination in air, a possible solution to this problem is to increase the spring constant of the cantilevers used in VCM to overcome the capillary force. From the following experiment, we can see that the JTC is eliminated with cantilevers of sufficiently large spring constant.

Before discussing the experiment, we should describe the JTC process in air. A sample surface in ambient air is always covered with a layer of surface contamination, which is believed to be "liquid-like" matter.⁸¹ When the tip begins to enter this layer, a meniscus will form and an attractive capillary force will be applied to the tip. From equation (3.18), we can derive that the capillary force gradient has a finite value. If the spring constant of the cantilever is smaller than this force gradient, the deflection of the cantilever can never balance the attraction force. In this case, the tip will be continuously accelerated towards the surface until tip-sample contact, which is the JTC

process. It has been claimed that the tip will always be pulled to the surface by this capillary force.⁸ If this indeed happens, we should always see the JTC. To test this conclusion was the original motivation of measuring loading curves for cantilevers of different spring constants.

Three silicon cantilevers of different spring constants were used. The cross-section of these cantilevers is trapezoidal, with the top side wider and the bottom side (where the probe is mounted) narrower. Their spring constant, k , is calculated from the geometry⁷⁶

$$k = \frac{1}{8} E(w1 + w2) \frac{t^3}{L^3}, \quad (5.7)$$

where E is the Young's modulus (for silicon $E = 1.79 \times 10^{11}$ N/m²), L is the length of a cantilever, t is the thickness of a cantilever, and $w1$ is the width of the bottom side and $w2$ is the width of the top side of a cantilever. The geometric dimensions were measured with the SEM.

Table 1. Parameters of the three silicon cantilevers used for the loading curve comparison.

Model No.	Tip No.	$L(\mu\text{m})$	$t(\mu\text{m})$	$w1(\mu\text{m})$	$w2(\mu\text{m})$	k (N/m)
1620	#3	450	4.20	18.00	37.70	1.013
1660	#8	220	6.50	20.00	55.00	43.28
1650	#25	120	3.95	28.00	47.00	59.85

Figure 5.8 shows the results of loading curves for these three silicon cantilevers. The sample is an aluminum coated optical mirror whose low surface roughness helps reduce the influence of surface topography. Loading curves were measured on the vibration isolation stage to minimize vibration noise. Figure 5.8(a) shows obvious tip jumping in both the approaching and pull-back curves. In Figure 5.8(b), the tip responds to the attractive interaction in a controlled manner. The response to the attractive force in Figure 5.8(c) is negligible.

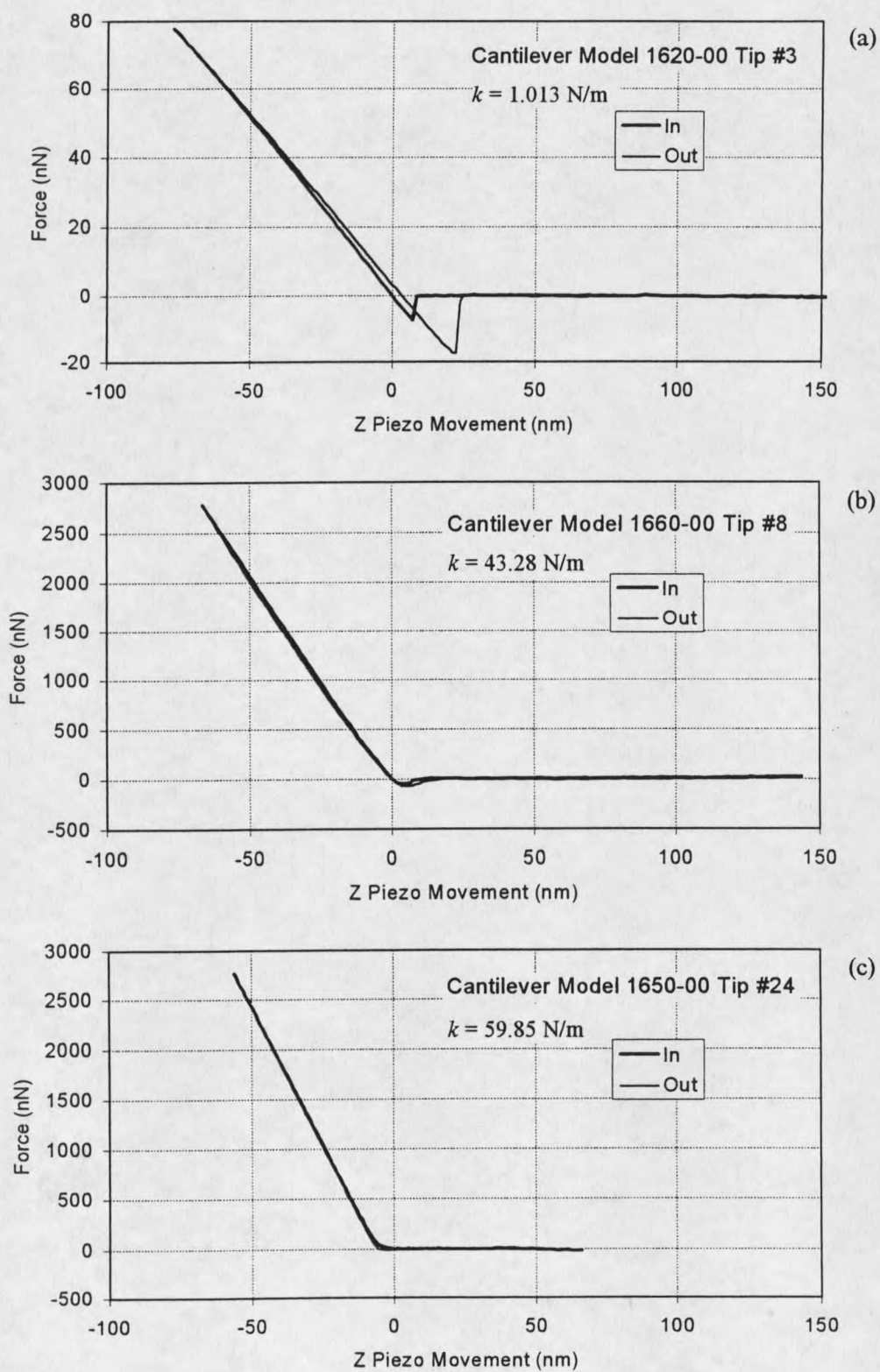


Figure 5.8. Loading curves for cantilevers of different spring constants.

Figure 5.9 is an enlarged view of the three approach curves in Figure 5.8, which shows the cantilever deflection around the tip-surface contact point. We see that for the two cantilevers of greater spring constants, JTC does not occur. This indicates that in an ambient environment with a sufficiently large spring constant, cantilever control can be maintained and only the controllable ramp-to-contact (RTC) can occur. That means a tip can be brought to very close proximity without being pulled to contact with the surface, and thus the assumption that the tip will always be captured by the surface contamination layer is incorrect.

From the comparison of those three cantilevers, we can see that the method of solving the JTC problem is to use cantilevers that have a higher spring constant, since the gradient of the attractive force that pulls the tip toward the sample is finite. If we use a cantilever that has a spring constant greater than the maximum force gradient of the attraction, JTC can be prevented. With a cantilever of a large spring constant that is greater than the maximum attractive force gradient, the tip can stay very close to the surface within the contamination layer, and thus remains out of contact with the surface.

Surface Contamination Studies

In ambient air, the ever-present layer of contamination existing between the tip and the sample is one of the most important factors that govern the tip-sample interaction. Loading curve measurement is a direct method to probe this contamination layer. Under vacuum most of the contamination is removed from the surface, particularly the liquid-like layer is removed, and hence comparing the loading curve in air and in vacuum can provide information about this contamination layer. Specifically, an estimation of the thickness of surface contamination and the magnitude of capillary attraction can be made with the loading curve measurement. In this connection we have carried out a series of experiments to obtain such information.

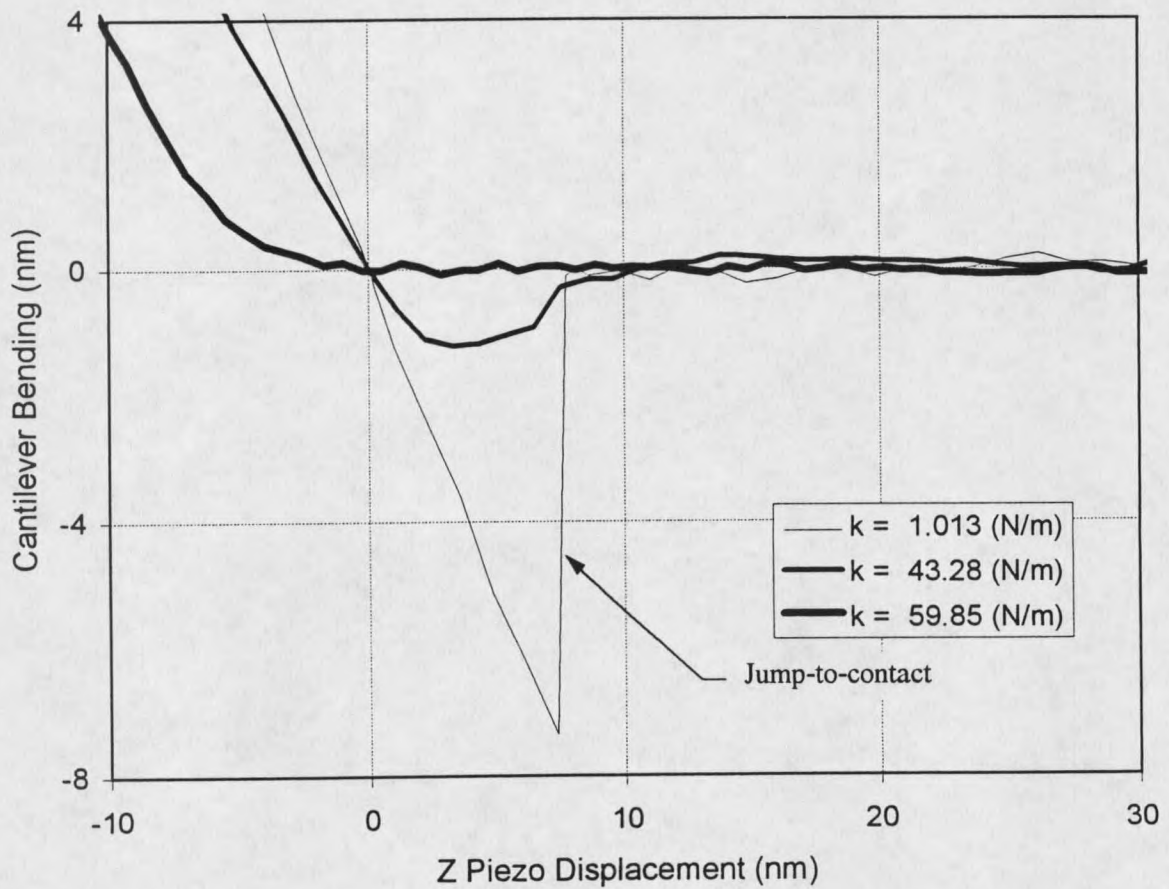


Figure 5.9. Comparison of the approach curves in the loading curves of three cantilevers with different spring constants near the tip-surface contact point.

To minimize the effect of surface topography on the loading curve measurement, a gold sample with surface roughness of about 10 nm was used. This sample surface, that had been exposed to air for more than two months, presents a stable surface contamination layer. The tests were performed with a silicon tip, model No. 1660-00 #8, in the main chamber on the vibration isolation stage. First the loading curve measurement was performed in air, then the chamber was evacuated to about 1×10^{-6} Torr, and the measurements were again performed on the same area. Figure 5.10 shows one set of the experimental results.

From Figure 5.10, we can see that the jump-to-contact (JTC), which is the abrupt change in tip-sample gap, does not occur for this cantilever. This is desirable because we do not have any unstable states during the loading curve measurement. On the approaching curve measured in air, labeled "In" in Figure 5.10(a), we can see that the attractive force starts to become significant in the range of about 10 nm. On the other hand, for the approach curve measured in vacuum (Figure 5.10(b)), the attractive force is much weaker.

Comparing these two approaching curves, we can see that the attractive force that was detected in air by this cantilever is mostly the result of the capillary force of the surface contamination layer, rather than the "non-contact" type interaction between the surfaces of the tip and the sample. Therefore, we can use the theoretical model to fit the approach curve measured in air to get an estimation of the thickness of the contamination layer.

Figure 5.11 shows the result of the fitting, which gives the initial tip-sample gap where the capillary force starts to appear as $G_0 = 11.6$ nm. Under ideal conditions, this is equal to the total thickness of the contamination layers on both the sample and the tip. We shall assume the thickness of the contamination layer on the tip and sample surfaces to be the same. Therefore, the thickness of the surface contamination layer can be estimated to be about 6 nm. From Figure 5.11 we can also obtain the magnitude of the maximum capillary force, for this particular experiment, to be about 230 nN. This measured capillary force magnitude is consistent with the values measured by other researchers.⁵

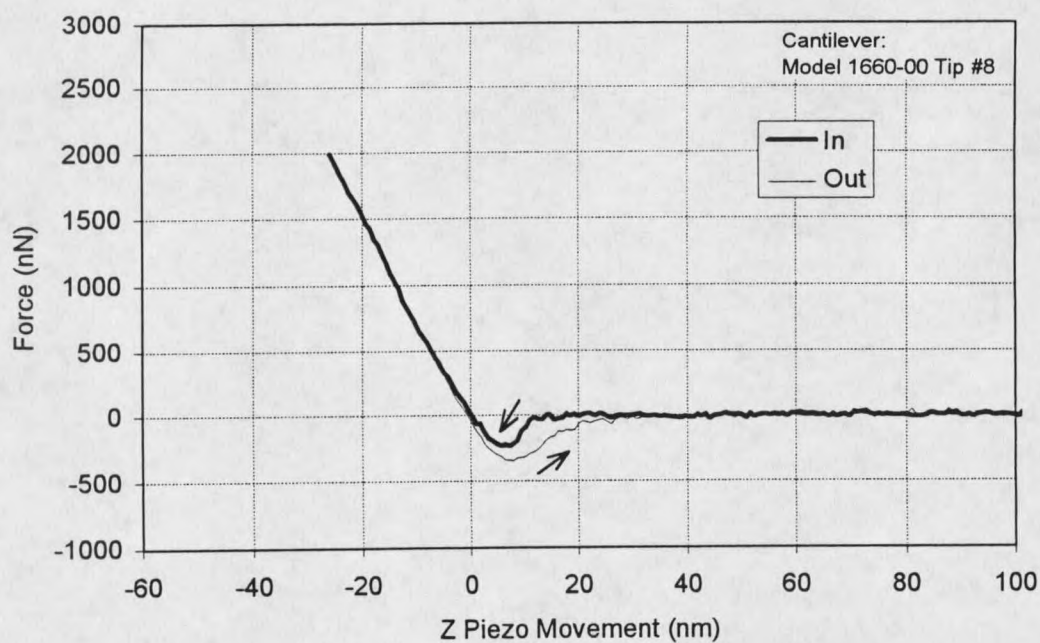
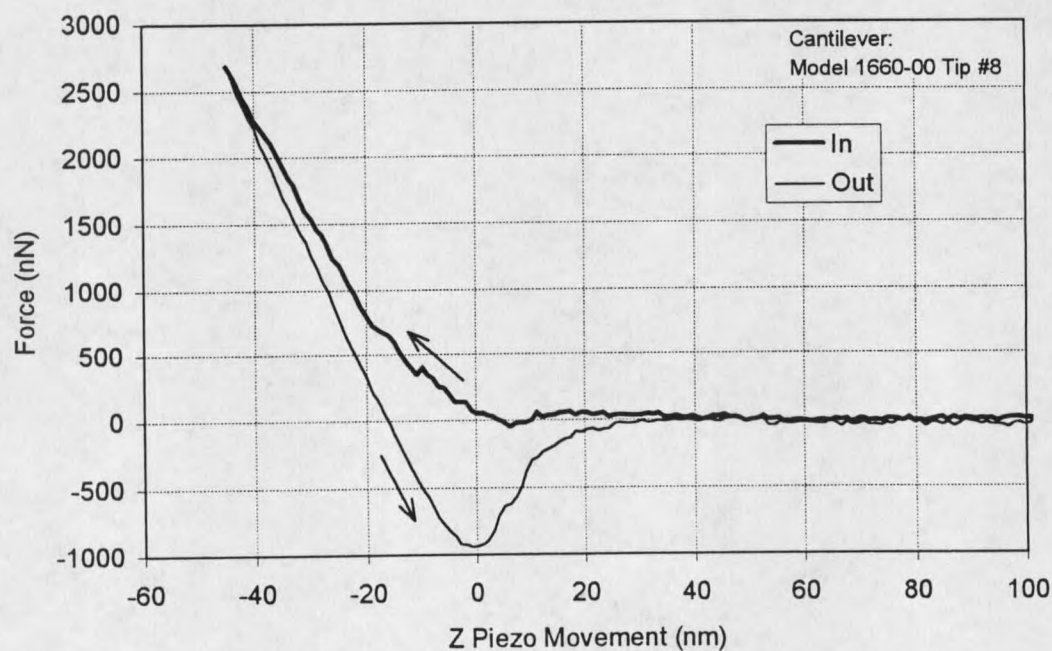
(a) in air**(b) in vacuum**

Figure 5.10. The loading curves measured in (a) air and in (b) vacuum with the same cantilever on the same sample area.

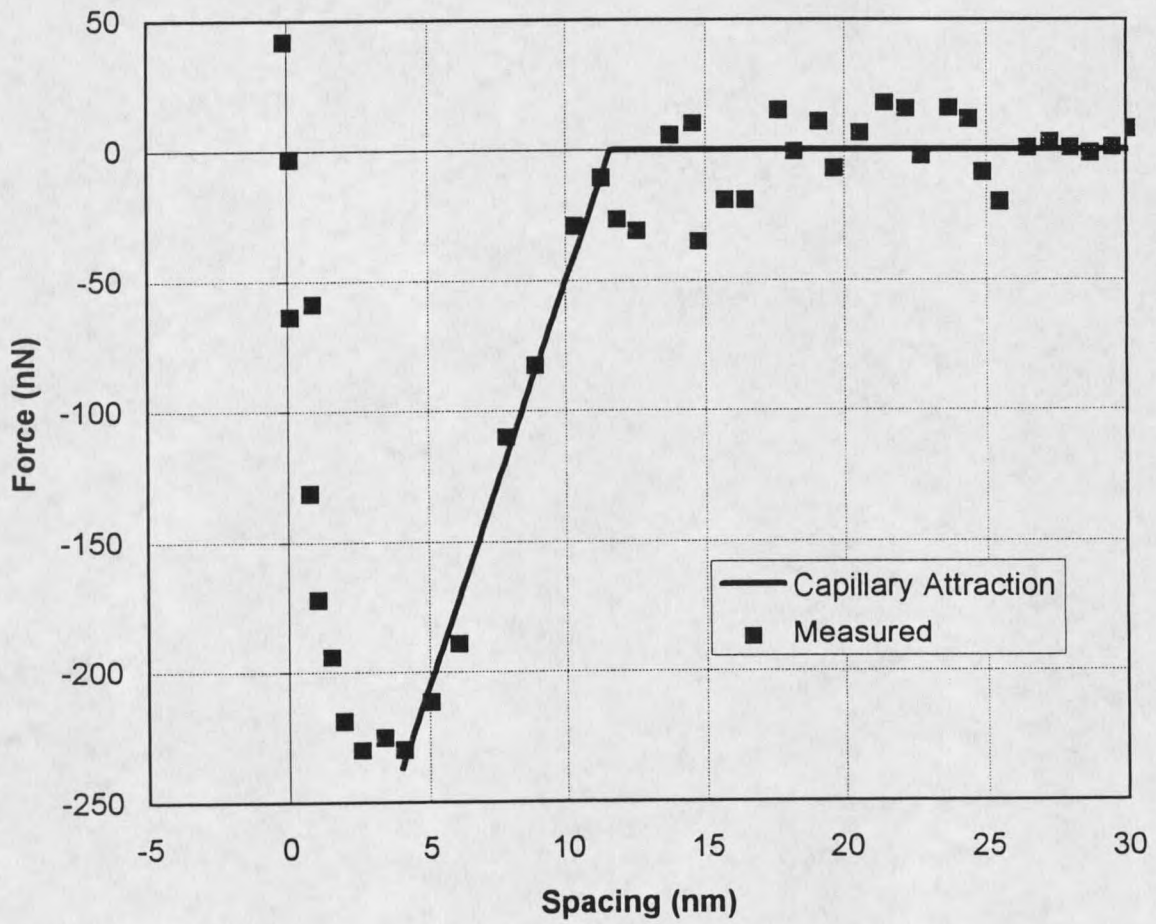


Figure 5.11. Fitting the approach curve with simplified model of capillary attraction force. $R = 800$ nm, $G_0 = 11.6$ nm.

Conclusion

The loading curve measurement is an important method in probing tip-sample interaction and surface contamination layers. An analysis method of processing the experimental loading curves is introduced, which is called cantilever-deflection correction. A raw experimental loading curve involves cantilever deflection for the force measurement, which does not show the true tip-sample gap. To plot the loading curve as a function of tip-sample gap for comparison with theoretical models, the effect of change in tip-sample gap due to cantilever deflection must be removed from the loading curve.

Since the loading curve measurement involves tip-sample contact, a number of uncertain factors make the reproducibility of the measurement poor. We performed systematic studies on the reproducibility issue by varying contact location, sample material, AFM tip, and environment. Since the contamination layer is mostly removed under vacuum, by comparing the loading curves measured in air and in vacuum, some properties of this layer can be studied. By comparing the results obtained in air and under vacuum, we found that the contamination layer is a major factor contributing to the fluctuation of loading curve measurements. It has hitherto been believed that the capillary attraction due to the surface contamination layer will always capture the AFM tip in the JTC process, thus disabling VCM operation. By studying the JTC in the loading curves with cantilevers of different spring constants, we found that cantilevers with larger spring constants can resist the pulling of the capillary force of the surface contamination layer without being captured. Since the capillary force is the dominant attractive force in air, by fitting the approach curves obtained in air with the theoretical model of capillary force, we obtain an average value of the contamination thickness to be about 6 nm.

CHAPTER 6

STUDIES OF VIBRATING CANTILEVER MODE

Introduction

The vibrating cantilever mode (VCM) was demonstrated as early as the first AFM paper¹. In this mode of AFM operation, the cantilever is set to oscillate mechanically on or near its resonant frequency. When the tip is brought to the surface and if the tip-sample interaction is large enough, significant changes will occur in the resonance characteristics, such as the resonant frequency and quality factor of the cantilever. By detecting the amplitude change or phase shift of the cantilever vibration, the sample surface can be probed for topographic imaging. Theoretically, the change in resonance characteristics takes place before the tip and the sample come into contact, which means that the surface can be probed without physical contact. For this reason, the VCM has been called the non-contact mode.⁸⁴

Although the VCM operation in AFM has been investigated since 1986, a number of issues remain. First, for examination of samples in air, the surface contamination layer complicates the meaning of non-contact, contact, etc. The imaging mechanism of a vibrating tip interacting periodically with a contaminated sample surface is not clear. Second, the VCM has more control parameters than the DC (that is, the continuous contact) mode, such as the resonant frequency, the operation frequency, driving amplitude, etc. The relationship between these control parameters and their influence on the image quality is not clear. Finally, the possibility of tip and sample damage under various conditions has not been examined. In reality, high resolution, stable VCM image has not been routinely obtained. The goal of these studies is to understand the image mechanism and to

develop an optimal operation method for obtaining maximum image resolution and best image quality while minimizing tip and/or sample damage.

In the traditional non-contact mode of operation, the amplitude of cantilever vibration is small (on the order of 1 nm). In the case of operation in air and with no static charges, the capillary force is the dominant interaction between the tip and the sample when the vibrating tip is brought close to the surface. While moving towards the sample, the tip experiences an increasing attractive force. This force gradient effectively reduces the spring constant of the cantilever, which lowers the resonant frequency of the vibrating cantilever, as described in equation (3.36). Such change in the resonant frequency can cause significant change in vibration amplitude and phase shift, which can be detected for establishing the topographic feedback to maintain the tip-sample gap at a constant. In this case, the tip is usually a few nanometers away from the sample surface due to the existence of a surface contamination layer. This limits the lateral resolution to a few nanometers because the lateral resolution is proportional to the tip-sample gap. In order to increase the lateral resolution, the tip must be brought closer to the sample. Since the thickness of the contamination layer is normally on the order of a few nanometers, the tip needs to work within the contamination layer to achieve the lateral resolution on the order of one nanometer.

One of the problems that is caused by the surface contamination layer is the capillary force as discussed on page 34. It can pull the tip towards the sample surface, which causes the tip to crash onto the sample, and results in capture of the tip on the sample surface. When this happens, the cantilever ceases to vibrate, and the VCM fails. In order to overcome this problem, operation modes with large vibration amplitudes have been developed.^{6, 8} When the VCM is operated with large vibration amplitudes, the tip moves in and out of the contamination layer. It has also been believed that in this mode the tip is brought to contact the surface periodically at the bottom of each oscillation cycle.⁸ Although the large vibration amplitude produces a high signal-to-noise ratio, which simplifies the operation, the stronger periodic tip-sample interaction may cause tip or

sample damage. The tip damage directly reduces the resolution of the AFM images because of the increase of the tip radius. Therefore, studying the conditions of the probe tips under different operations is important for developing the optimal operation conditions:

To search for an optimal operation condition of the VCM, a number of experiments were conducted with different operation parameters. After analysis of the experimental results, a new method of operating the VCM is developed in this thesis, that can routinely provide stable high resolution topographic images while minimizing tip and/or sample damage. This optimal operation method of VCM makes use of the final layer of surface contamination molecules to prevent direct physical contact between the tip and the sample. This newly developed operation mode is called the *near-contact mode*, because this new method maintains a minimal tip-sample gap on the order of angstroms. A theoretical model of the tip-contamination-sample system is proposed to explain the working mechanism of various vibrating cantilever operation modes. And it is demonstrated that the near-contact mode is the optimal operation mode.

Experiments

Experiment One: Changes in Tip Geometry by Periodic Contact

In the past, the condition of the tip of AFM probes after imaging in the periodic contact condition was not directly analyzed. This experiment is designed to study the changes of tip geometry after imaging a rough sample with two distinct sets of operation parameters. The first set maintains the tip-sample in the traditional non-contact mode, while the second set uses larger vibration amplitude, which results in periodic contact. The sample is an aluminum film with rough grain surface.

In this experiment, the AFM images were acquired with the sample in ambient air in the load-lock chamber of the combined AFM/SEM instrument and the SEM images of the AFM probes were acquired in the vacuum environment of the SEM main chamber with a vacuum pressure of about 10^{-6} torr. With the combined AFM/SEM instrument, the entire AFM including the sample

and probe can be moved between the load lock chamber and the main chamber with a mechanical transfer mechanism. The sample and probe are never removed from the AFM instrument, ensuring that any damage to the probe could be caused only during scanning the sample surface. The SEM images of AFM probes are acquired by rotating the AFM scanner 45 degrees from the position where AFM images are acquired.

A series of measurements with a single probe were performed on the same aluminum grain sample. A silicon VLS probe (IBM model number VLSHF125) with a 359 kHz resonant frequency was used, whose resonance spectrum is shown in Figure 6.1. The Q-factor of this cantilever is about 250. Extreme care was taken while making these measurements to ensure that the tip was not damaged by touching the sample when the AFM was moved between the SEM chamber and the load-lock chamber or during tip approach to the sample. Each tip approach was performed in a similar manner by monitoring the decrease in cantilever vibration amplitude. The same feedback control parameters were used for all of the AFM images. The following procedure was followed:

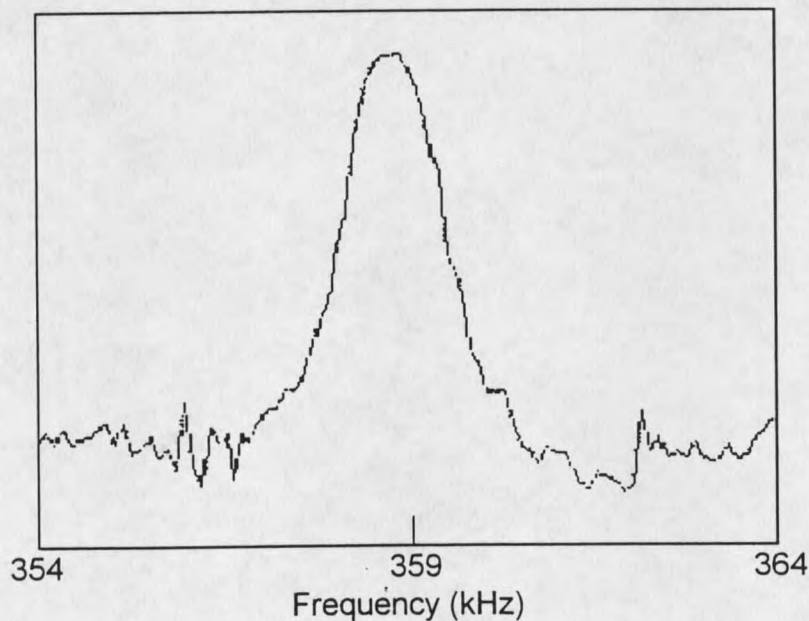
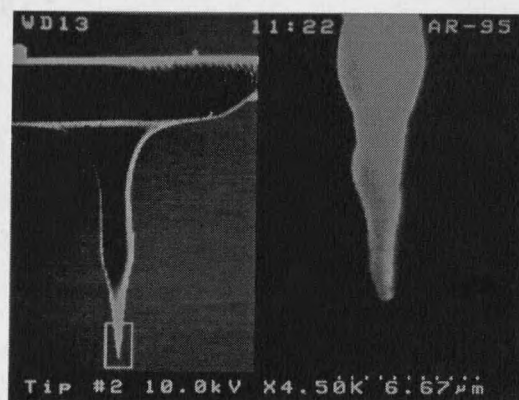


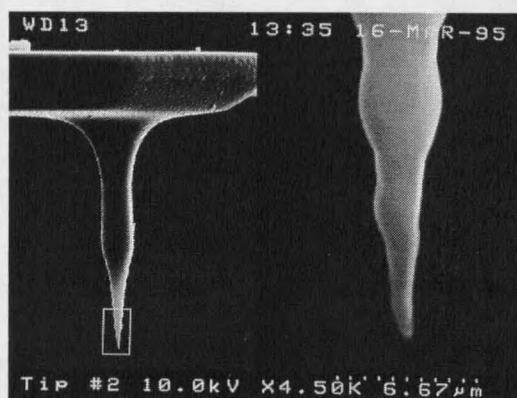
Figure 6.1. The resonance spectrum of the VLS silicon probe.

- 1) The sample and AFM probe were mounted in the AFM/SEM and a SEM image of the probe tip was acquired, which is shown in Figure 6.2(a).
- 2) The AFM head was then moved to the load lock chamber and small amplitude AFM images were acquired for a period of two hours. The operation parameters are: free cantilever vibration amplitude of 8 nm, and the feedback level is based on a 40 % reduction in vibration amplitude. Figure 6.3(a) illustrates a typical acquired topographic image.
- 3) The AFM head was moved to the SEM chamber and a SEM image of the probe was acquired which is shown Figure 6.2(b).
- 4) After the AFM was moved back to the load lock chamber, an AFM image was measured with the conditions described in the step 2) above. It was verified that the AFM image appeared qualitatively the same as shown in Figure 6.3(a).
- 5) The final image was acquired with a large vibration amplitude. The operation parameters are: free cantilever vibration amplitude equals 80 nm, with a 10 % reduction for feedback. The resultant AFM image is presented in Figure 6.3(b).
- 6) Finally, the AFM head was transferred to the SEM and the tip image, shown in Figure 6.2(c), was measured.

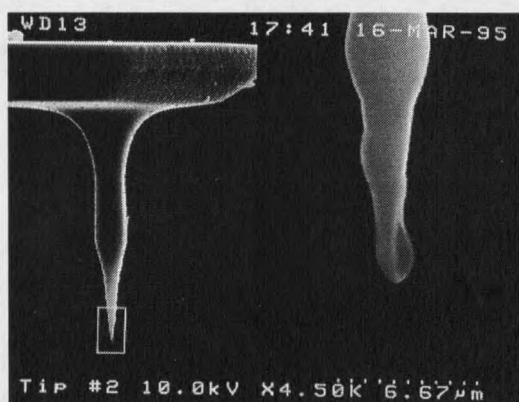
From Figure 6.2, a change in the tip geometry was found where the large amplitude operation made the tip blunt. Comparison of Figure 6.2(b) with Figure 6.2(c) clearly indicates a dramatic change in tip geometry caused by imaging with large vibration amplitude. An 80 nm cantilever vibration and a 10% reduction in vibration amplitude are the conditions reported to cause repeated contact between the probe tip and sample⁸. The change in probe geometry appears to be the result of contamination build-up on the probe during the repeated tip-sample contact process.



(a)



(b)



(c)

Figure 6.2. SEM image of the VLS silicon tip. (a) Before any topographic AFM image was acquired, (b) after three hours of imaging with a small (8 nm) vibration amplitude with a 40% reduction in amplitude, and (c) after one topographic image with 80 nm vibration with a 10% reduction in amplitude. The scale dots on the lower right corner is for the image in the left. The right image corresponds to the box in the left image. Its scale is 1/10 of the left image.

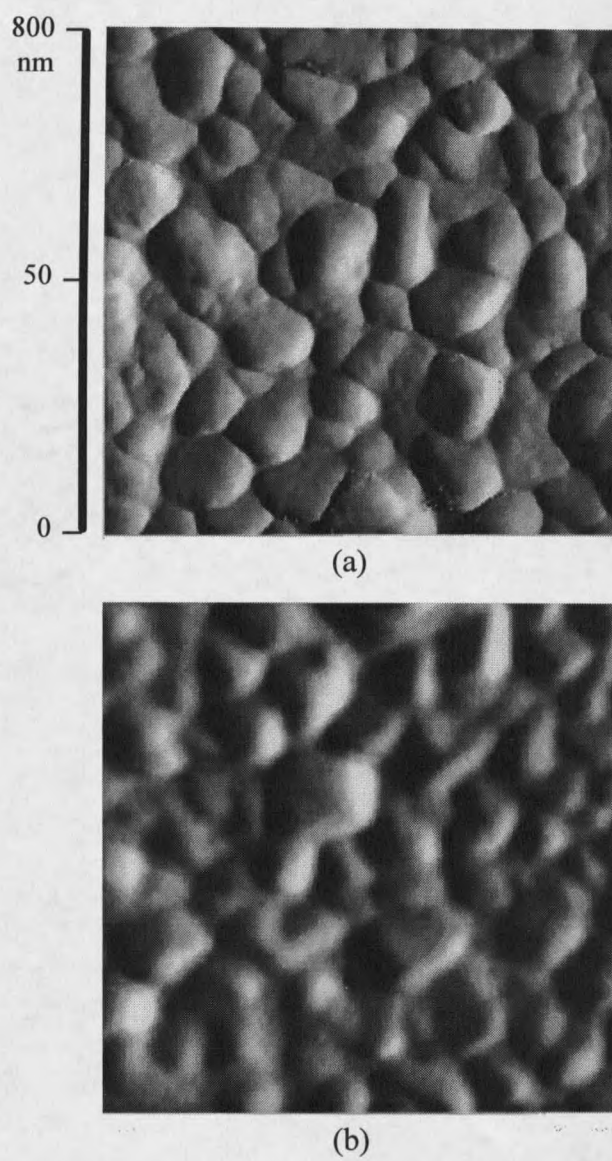


Figure 6.3. Topographic images (shaded view) of aluminum grains: (a) With 8 nm vibration amplitude and (b) with 80 nm vibration amplitude. The size of both images is 800×800 nm.

Visual inspection of the AFM images shows that the image acquired by large amplitude operation is not as sharp. Figure 6.3(a) and Figure 6.3(b) appear substantially different. The grains in Figure 6.3(a) have surface detail that is not observed in Figure 6.3(b). To get a quantitative comparison of the lateral resolution, we can perform Fourier transform to the scan lines of the topographic data, which results in a spatial frequency spectrum of the AFM image. A similar technique has been used to analyze SEM images⁸⁵ where a two-dimensional Fourier transform was used to create a frequency space image. Since the small features in an image give the high frequency components in its spatial frequency spectrum, the density of the spatial frequency spectrum can be a measurement of the sharpness of the image, which is related to the lateral resolution.

For two images with the same noise level, the one that has the higher resolution would have a higher density in the higher frequency region. The difference in lateral resolution for the AFM images shown in Figure 6.3 can be quantified from their spectral analysis (Figure 6.4). The spatial frequency spectrum of the small amplitude AFM image (Figure 6.3(a)) has considerably larger high frequency components than the spectrum from the large amplitude AFM image (Figure 6.3(b)). This indicates that the AFM image with smaller cantilever vibration amplitude has better lateral resolution.

According to the SEM images, there is no apparent change in the geometry of the AFM probe tip before and after the small amplitude AFM imaging. Additionally, the AFM images measured using the 8 nm vibration showed no change in clarity over two hours of imaging. Because the vibration amplitudes of the probe is similar to the thickness of the surface contamination layer, the vibrating tip is maintained in the contamination layer. If the probe tip were not entering the contamination layer we would expect to observe a decrease in resolution caused by hydroplaning of the tip over the contamination layer.

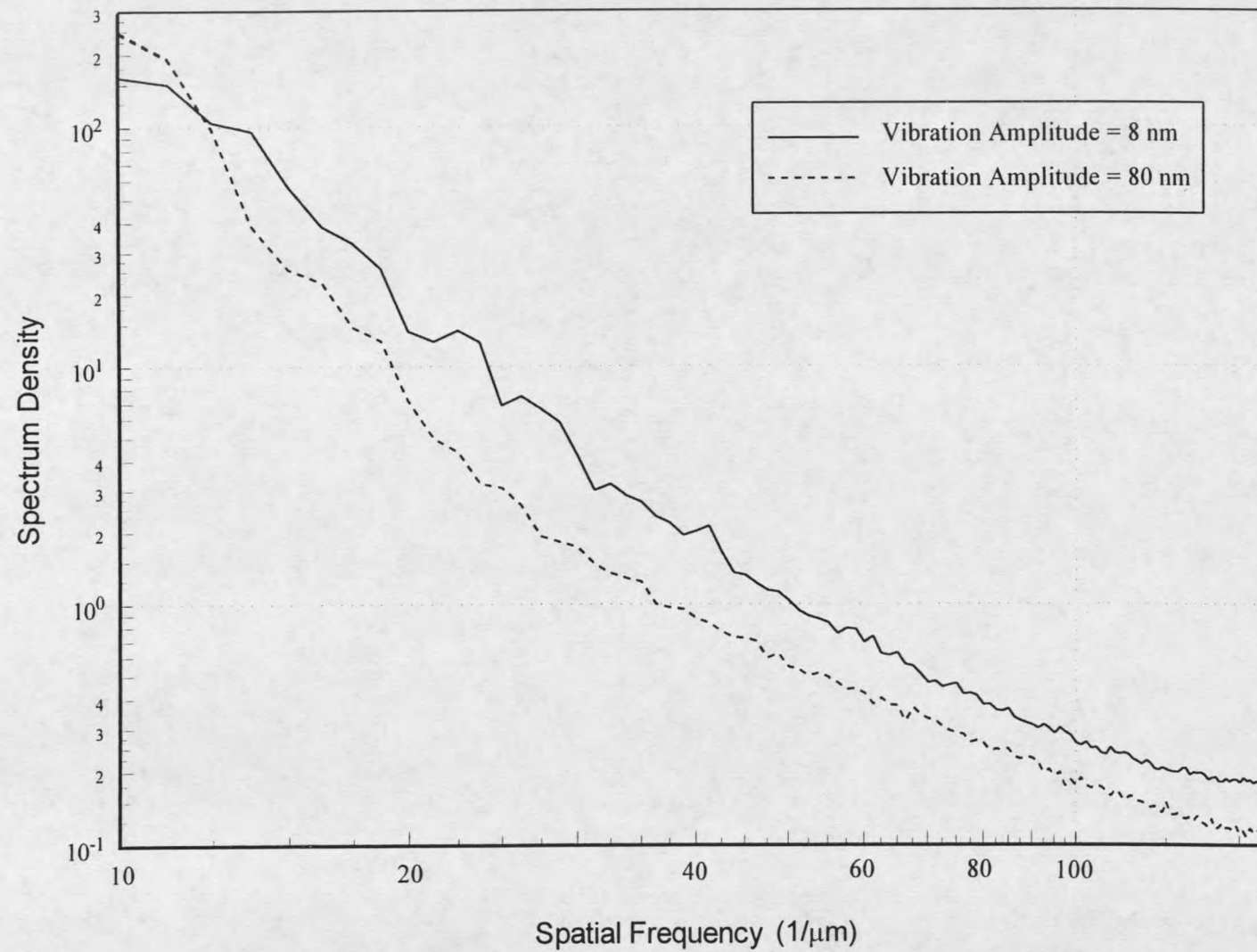


Figure 6.4. Spatial frequency spectrum of the topographic images illustrated in Figure 6.3.

Experiment Two: Wear of a Silicon Tip on a Silicon Sample

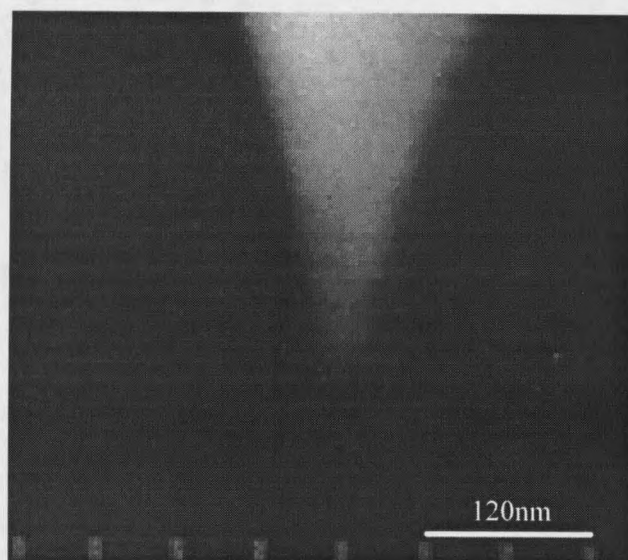
It is known that physical contact between two objects of the same material can result in "vacuum welding". When these two objects that are "welded" together are separated, there would be atom transfer between them. In the case of a silicon tip contacting a silicon sample, the atom transfer means tip and sample damage. With the help of SEM, we have the opportunity of investigating such a phenomenon. This experiment is designed to set the AFM operating in the periodic contact mode to have periodic physical contact between the tip and the sample.

A series of images of a silicon wafer was measured using a silicon probe, TopoMetrix model #1650-00. The cantilever vibration amplitude used for these images is 100 nm. The feedback was established with a 10% reduction in vibration amplitude. SEM images of the probe before and after measuring AFM images for a period of one hour are illustrated in Figure 6.5.

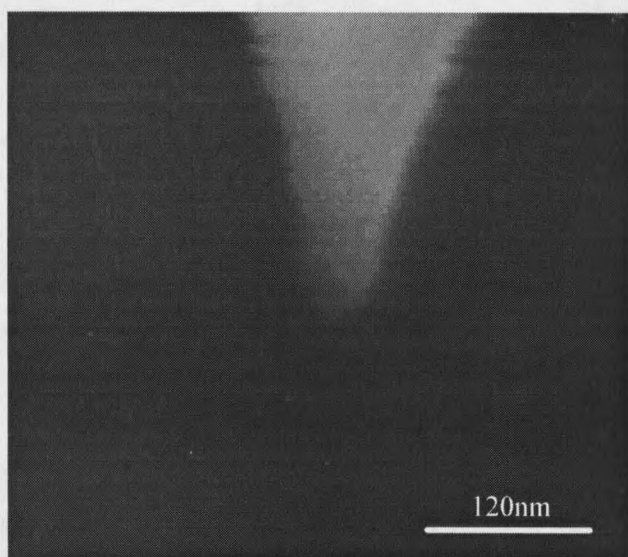
Comparing Figure 6.5 (a) and (b), it is clear that the tip was substantially changed while imaging the sample. The estimated radius before scanning is 10 nm. It is 20 nm after scanning. The change in tip geometry indicates that atom transfer between the tip and the sample does happen. This also confirms that the periodic contact condition does occur for the VCM operating at 100 nm vibration amplitude with feedback established at 10 % decrease in amplitude.

Experiment Three: Surface Modification by Periodic Contact

In the previous experiments, deformation of the tip profile is observed. If the tip-sample interaction is strong enough to cause this, it is possible that the sample surface is deformed due to the periodic contact process. To enhance the surface modification by the VCM imaging, a soft sample is preferred. In this experiment, the sample used is a thin polyimide coating on a glass substrate. Phase detection is used in this experiment.



(a)



(b)

Figure 6.5. SEM image of a silicon tip (a) before imaging a silicon surface and (b) after imaging with an oscillation amplitude of 100nm and a 10% reduction in amplitude.

Figure 6.6 shows the experimental process and the mechanism of surface modification. At first, a near-contact VCM image with vibration amplitude of 6 nm was acquired (Figure 6.7a). Then the probe was pulled out of feedback, and set to vibrate at an amplitude of 100 nm, and brought to feedback again. Once in feedback, the probe is operating in the periodic condition and sample modification is possible. The feedback set point is set to have just enough phase shift to bring the probe into feedback, to insure that minimal contact force is applied to the sample. The sample was scanned with the periodic contact mode in a range of 100×50 nm. Then the probe was pulled out of feedback, and the system was reset for near-contact imaging and a near-contact VCM image in the same area was acquired (Figure 6.7b). The same cycle was repeated once to make a periodic contact scanning on a large area (250×200 nm). The result of the periodic contact scanning is also recorded with the third near-contact image (Figure 6.7c).

For all the scans, scan direction is horizontal and starts from the top of the image. From Figure 6.7b and Figure 6.7c, obvious sample modification can be observed. Because of the particular scanning orientation, the "foot print" of the periodic contact imaging is more obvious on the left, the right, and the bottom of the rectangular scan area. This set of images clearly shows the sample damage by the periodic contact VCM operation.

The polyimide coating is a thin layer and its exact thickness is unknown. To get an idea of the thickness of this coating, the sample was imaged with the continuous contact mode AFM. In the first scan, the AFM image was very noisy, which indicates the instability of tip-sample interaction and the possibilities of sample damage. In the second scan acquired immediately after the first one, the image is stable and reveals the surface features of the glass substrate, which indicates that the coating is actually removed by the scanning in the continuous contact mode.

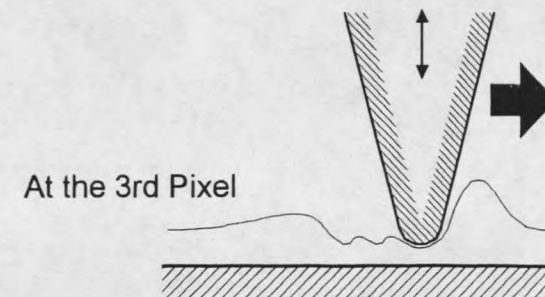
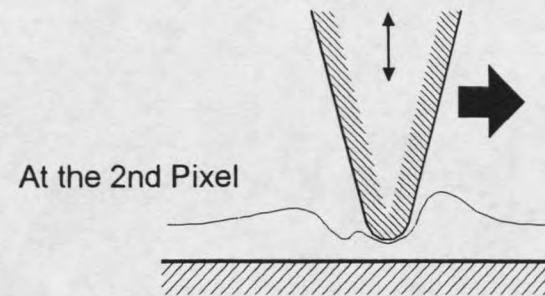
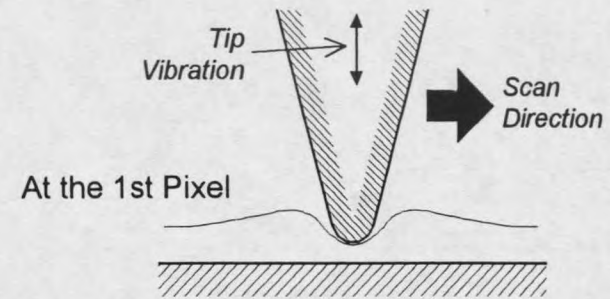
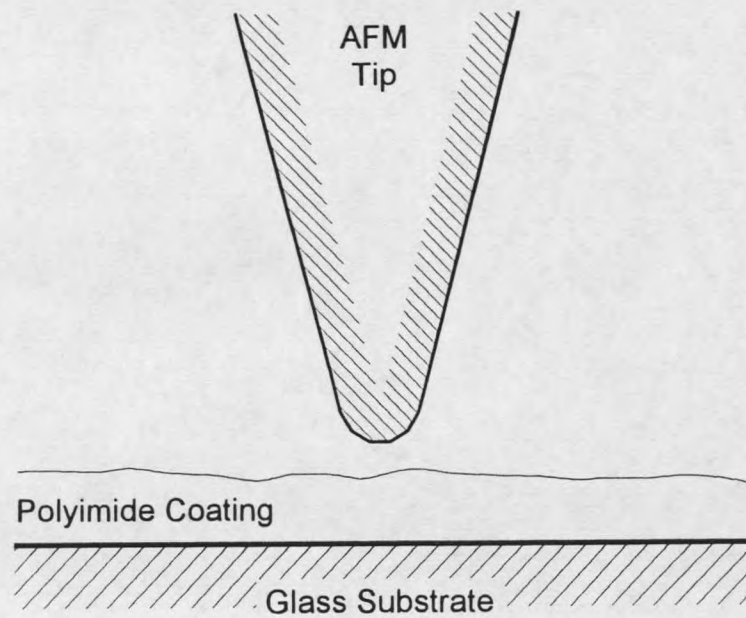
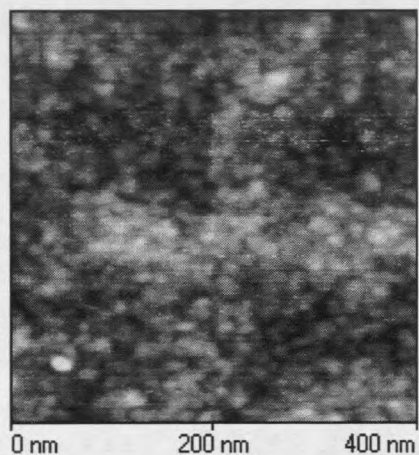
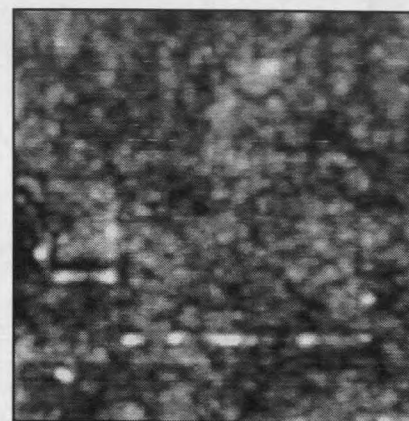
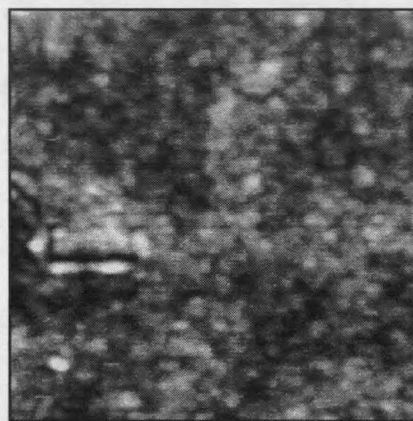


Figure 6.6. The mechanism of sample damage.

4.00 nm
0.00 nm



(a)



*The first periodic
contact scan area*

*Fast Scan
Direction*



*Slow Scan
Direction*



(b)

*The second periodic
contact scan area*



(c)

Figure 6.7. The near-contact AFM images (a)-(c) show that the sample is damaged by periodic contact imaging operations.

If the impact of the periodic contact is strong enough, the tip may break through the thin coating layer and interact with the glass substrate, which can result in tip damage. However, the SEM images (Figure 6.8) show no significant change in the tip profile, which indicates that the contact was light enough so that the polyimide coating was not broken through. Therefore, we can conclude that even when contacting the sample lightly, modification can take place on the surface of a soft sample like this polyimide coating.

Experiment Four: Monitoring the Tip Approach Process

Tip approach is an important step in PPM operation, as is mentioned on page 8. Monitoring the tip approach process can allow us to observe the different stages of tip-sample interaction and help design an optimal operation procedure.

The sample used in this experiment is a GaN wafer, the probe is TopoMetrix 1650-00 type, and the phase detection is used. First, the tip was brought into feedback with minimal change of the phase signal. Then the tip was gradually brought towards the surface by reducing the magnitude of the feedback setpoint. At each new feedback level, the phase signal was recorded for one second with a sampling rate of 1 kHz. The average value and the amount of fluctuation was monitored against the Z piezo movement, which is proportional to the tip sample gap.

Figure 6.9 shows an experimental result. The change in the fluctuation of phase signal (labeled "Average Deviation") clearly indicates the transition from the traditional non-contact to the near-contact region. The region that has large fluctuation is the operation region of the traditional non-contact mode, where the presence of the liquid-like contamination layer between the tip and sample causes the instability in the feedback loop, which results in large fluctuation in the phase signal.

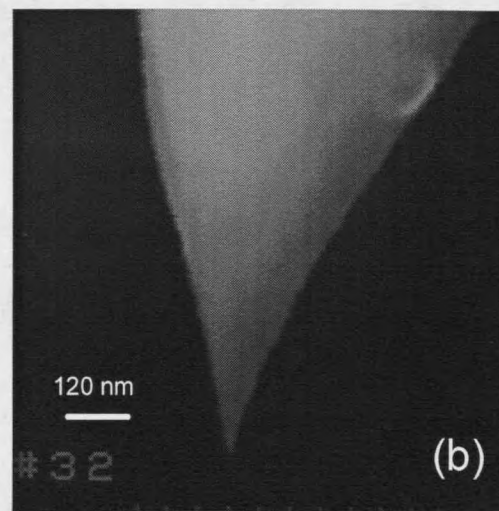
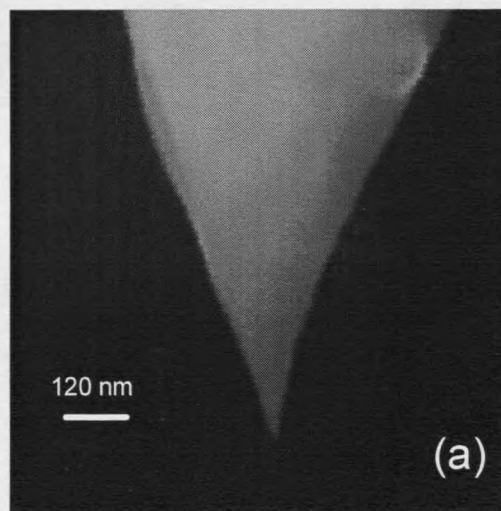


Figure 6.8. The SEM micrographs taken (a) before and (b) after the experiment (shown in Figure 6.7) show no obvious change in the tip profile.

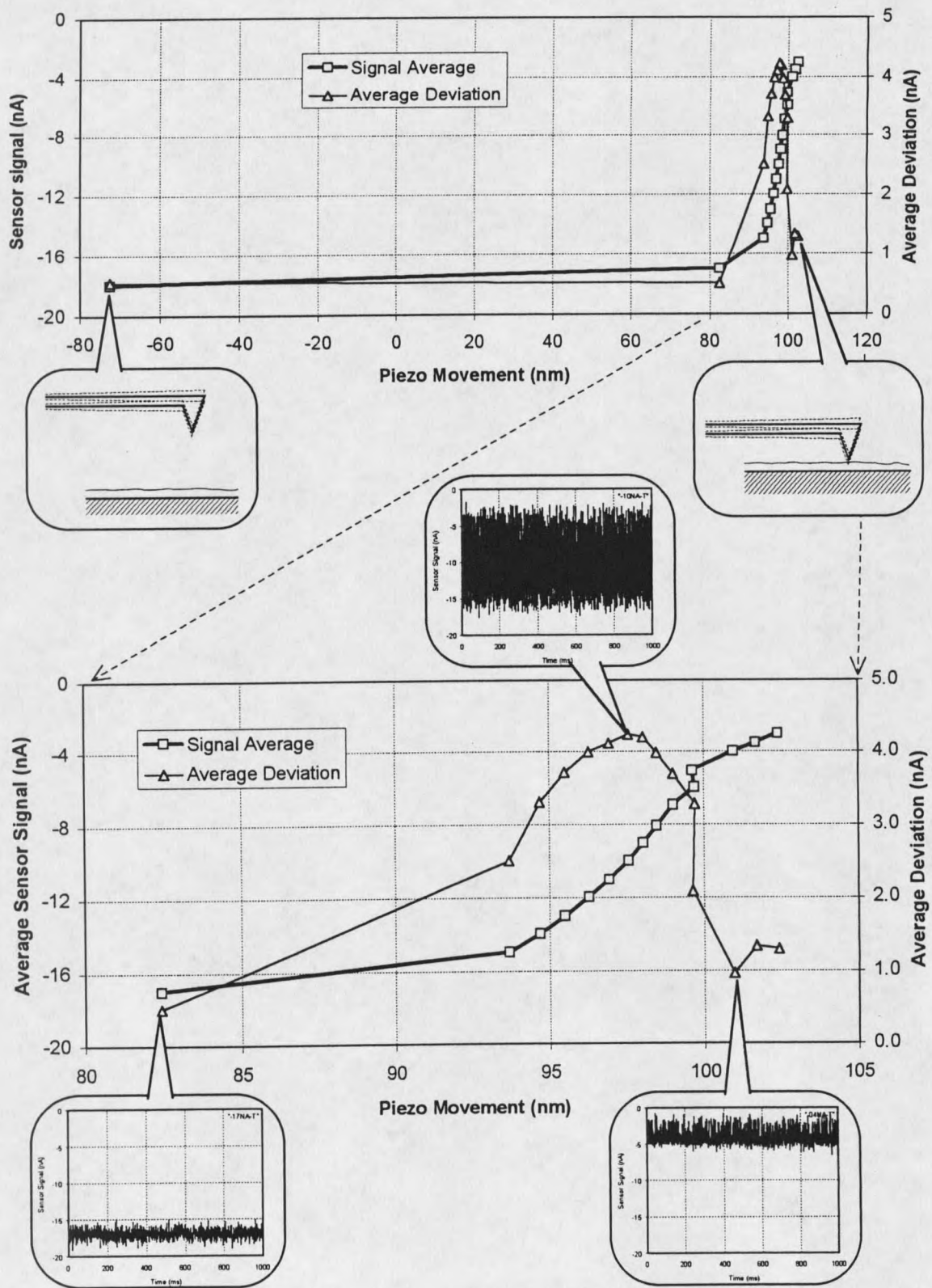


Figure 6.9. The transition from traditional non-contact to near-contact.

Experiment Five: Comparison of Three Distinct Operation Modes

This experiment is designed to compare the image quality generated by three different sets of operation parameters to determine the optimal VCM operating conditions. They represent the three distinct VCM operations: the traditional non-contact mode, periodic contact mode, and the new near-contact mode.

For this experiment, a polished alumina-silicate glass sample was used. This is a very good sample for testing lateral resolution. It has two kinds of surface features: the larger "cigar" sharp features on the order of 100 nm, and the smaller round features on the order of a nanometer. To maintain the original surface contamination layer, the sample was never in vacuum.

A silicon cantilever with $f_0 = 227.5$ kHz (Model 1650-00, #25) was used. Its resonance spectrum is shown in Figure 6.10. From the resonance peak, the Q-factor can be estimated to be about 400. Since the driving voltage was 4 mV when measuring this spectrum, and the piezo gain of the modulating piezo was 0.002 nm/mV, cantilever oscillation amplitude was about 3.2 nm on the resonance. The tip of the silicon probe was inspected with the SEM before and after the experiment. Three AFM images were acquired under three different conditions in the order listed in the following table.

Table 2. Experimental parameters of the three different VC operation modes.

	Piezo Amplitude (mV)	Cantilever Amplitude (nm)	Detection Mode	Feedback Setpoint	Operation Condition
(a)	3.4	3.0	Phase	67% change	Near-contact
(b)	3.4	3.0	Phase	12% change	Non-contact
(c)	136	108	Amplitude	10% change	Periodic contact

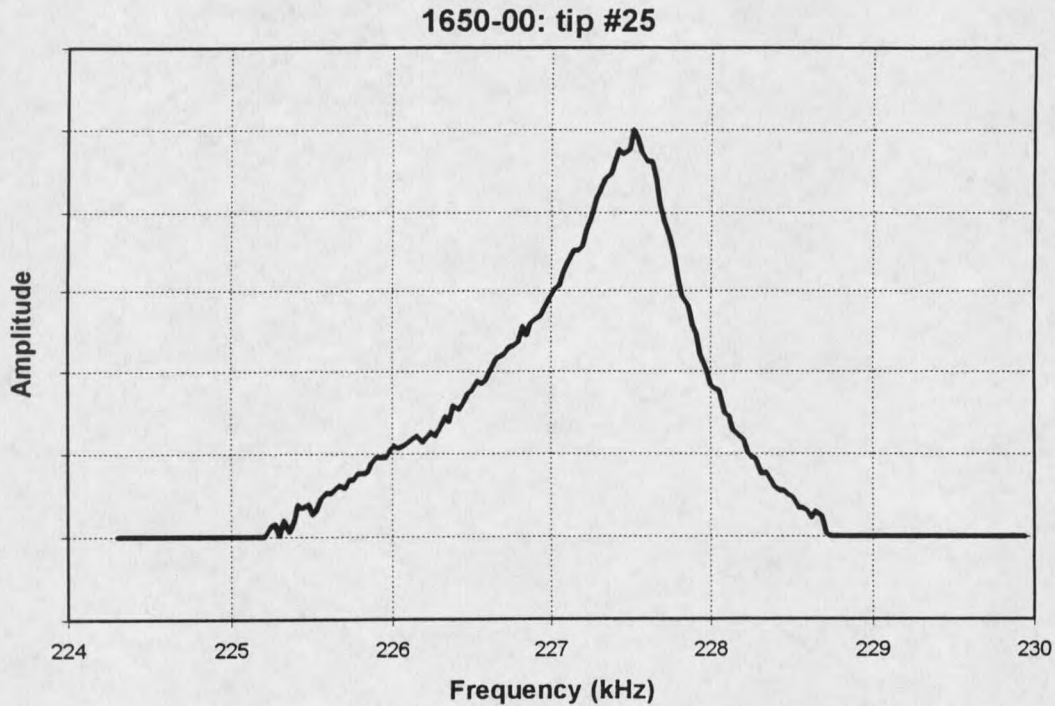


Figure 6.10. Resonance spectrum of the silicon tip (Model 1650-00 #25).

The AFM images are shown in Figure 6.11, where Figure 6.11(a), (b) and (c) were acquired with condition (a), (b) and (c) in the Table 3, respectively. The image sizes are 100 nanometers which is cut out of the original 1000 nm scan to show the same area on the sample surface. From these unfiltered images, we can see some significant differences. In the image acquired by the traditional non-contact mode, Figure 6.11(b), obvious ripples can be seen across the image, which is probably due to the instability of the surface contamination layer. The poor lateral resolution is also due to the tip being too far away from the surface. In the image acquired by the periodic contact mode, Figure 6.11(c), the image is very stable, since the periodic tip-sample contact eliminates the instability of the contamination layer. However, the lateral resolution is not very good because the sharpness of the tip is degraded by the periodic contact between the tip and the sample.

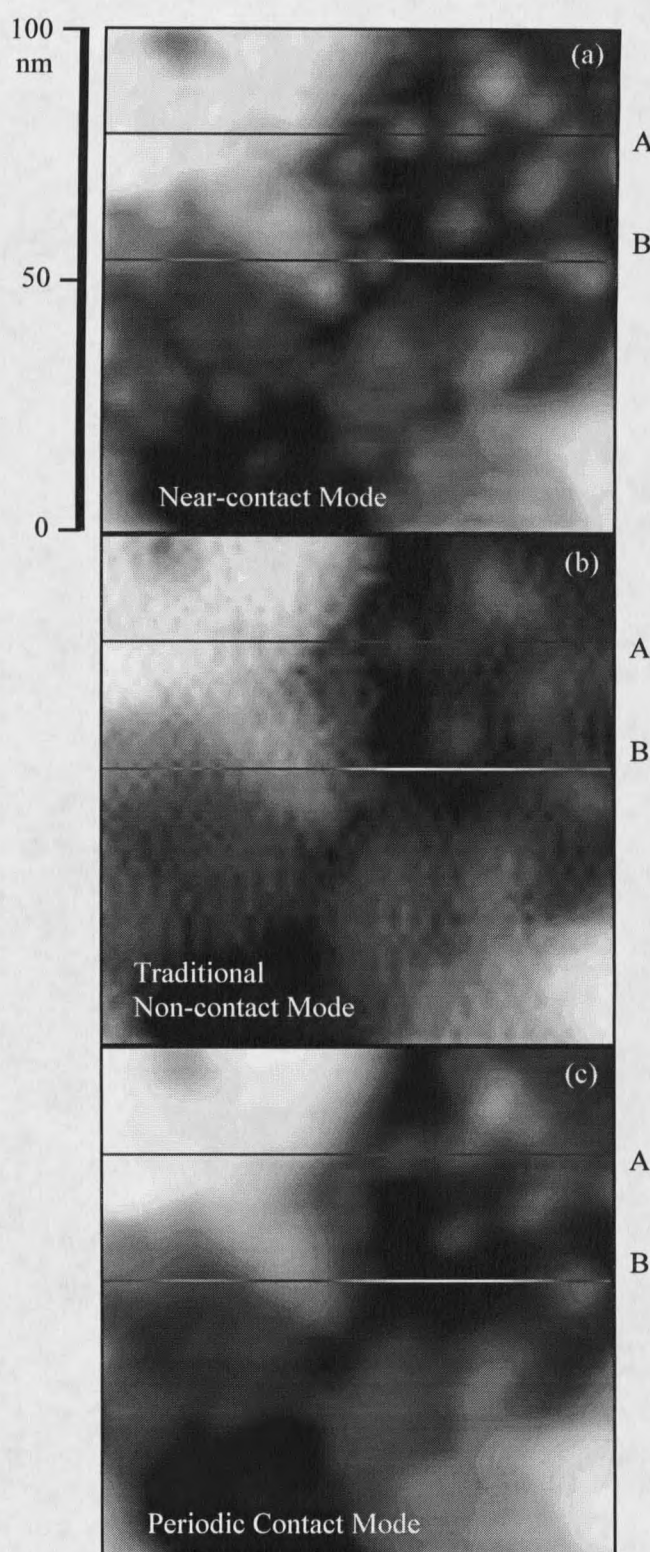


Figure 6.11. Topographic images of a polished alumina-silicate glass surface with three distinct operation modes. The image size is 100 nm $\times$ 100 nm.

In the image acquired by the near-contact mode, Figure 6.11(a), there also are no ripples because the near-contact mode can keep the tip very "near" the surface, so that the instability of the contamination layer can be removed. In this image, we can also see that the lateral resolution is significantly higher than the other two images. A number of small islands are resolvable in this image only because the near-contact mode can preserve the sharpness of the tip by preventing tip-sample contact while maintaining a very short distance between the tip and the sample.

To further illustrate the difference, two line profiles (A and B in Figure 6.11) across the same feature are extracted from those three images and displayed together in Figure 6.12. For all three images, the large features appears to be the same. Again, the ripples can be clearly seen on the scan lines from the traditional non-contact mode. In line A, peak α and β appear to be much sharper in the near-contact line. In line B, peak γ appears to be very sharp in the near-contact line. This peak also shows up in the non-contact line, although the height is similar to the size of the ripples, while it nearly disappears in the periodic contact line.

To get a quantitative comparison of the lateral resolution, we can perform Fourier transforms on the topographic data. Figure 6.13 shows the spatial frequency spectra of the same area of a 600×600 nm cutout from the original 1000 nm scan images. The near-contact image has substantially higher intensity in all frequency ranges compared to the periodic-contact image, which shows that the near-contact image contains more detailed information than the periodic-contact image. The spectrum of the non-contact image can be separated into two parts. First, for spatial frequencies smaller than $50 \mu\text{m}^{-1}$, the spectrum is almost the same as the spectrum of the periodic contact image. This means that images acquired by non-contact mode and periodic contact mode are the same for topographic features that are larger than 0.02μ . Second, for spatial frequencies larger than $50 \mu\text{m}^{-1}$, the spectrum density increases while the spatial frequency increases, which indicates that high frequency noise exists in the image. The spikes at about $120 \mu\text{m}^{-1}$ are due to the 60 Hz noise in the electronic system.

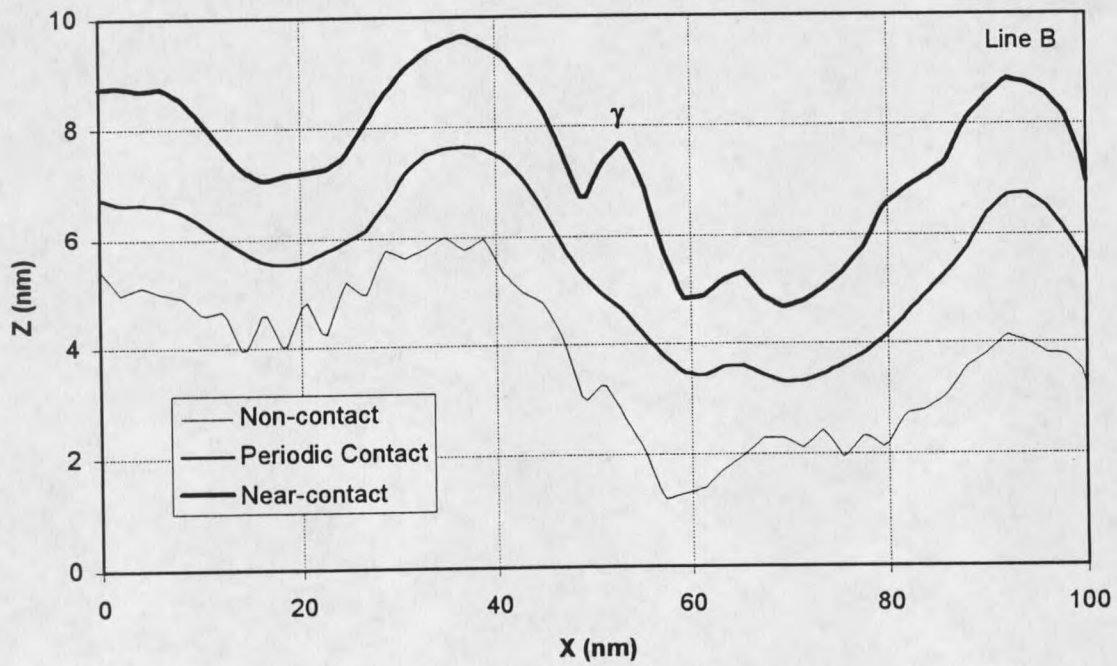
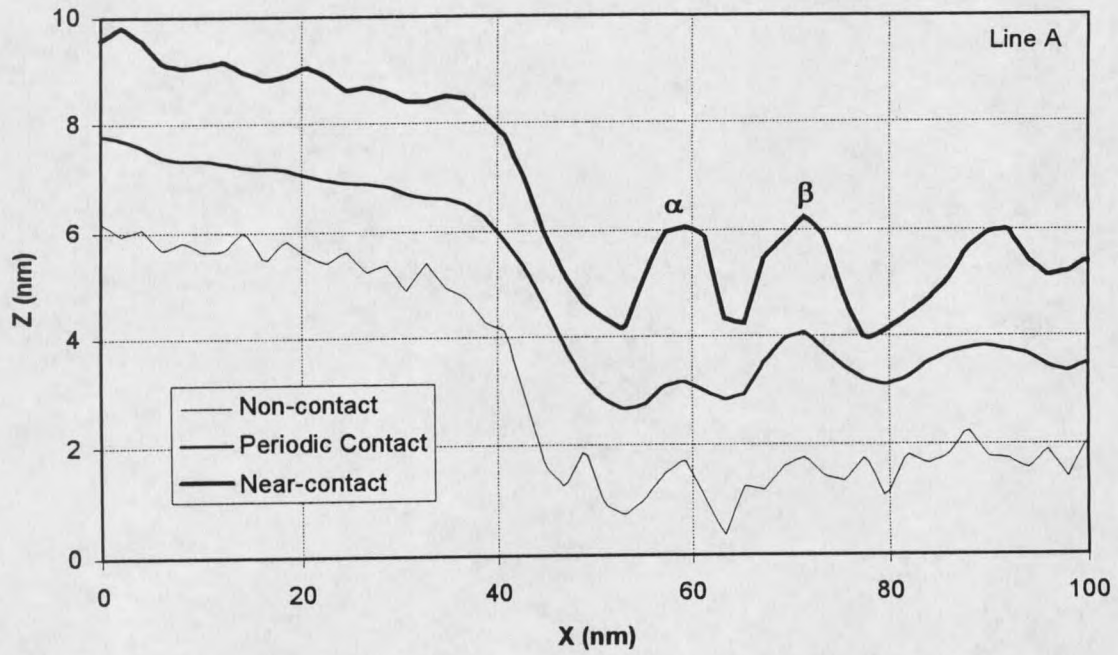


Figure 6.12. Line profiles of the same features by the three vibrating cantilever modes.

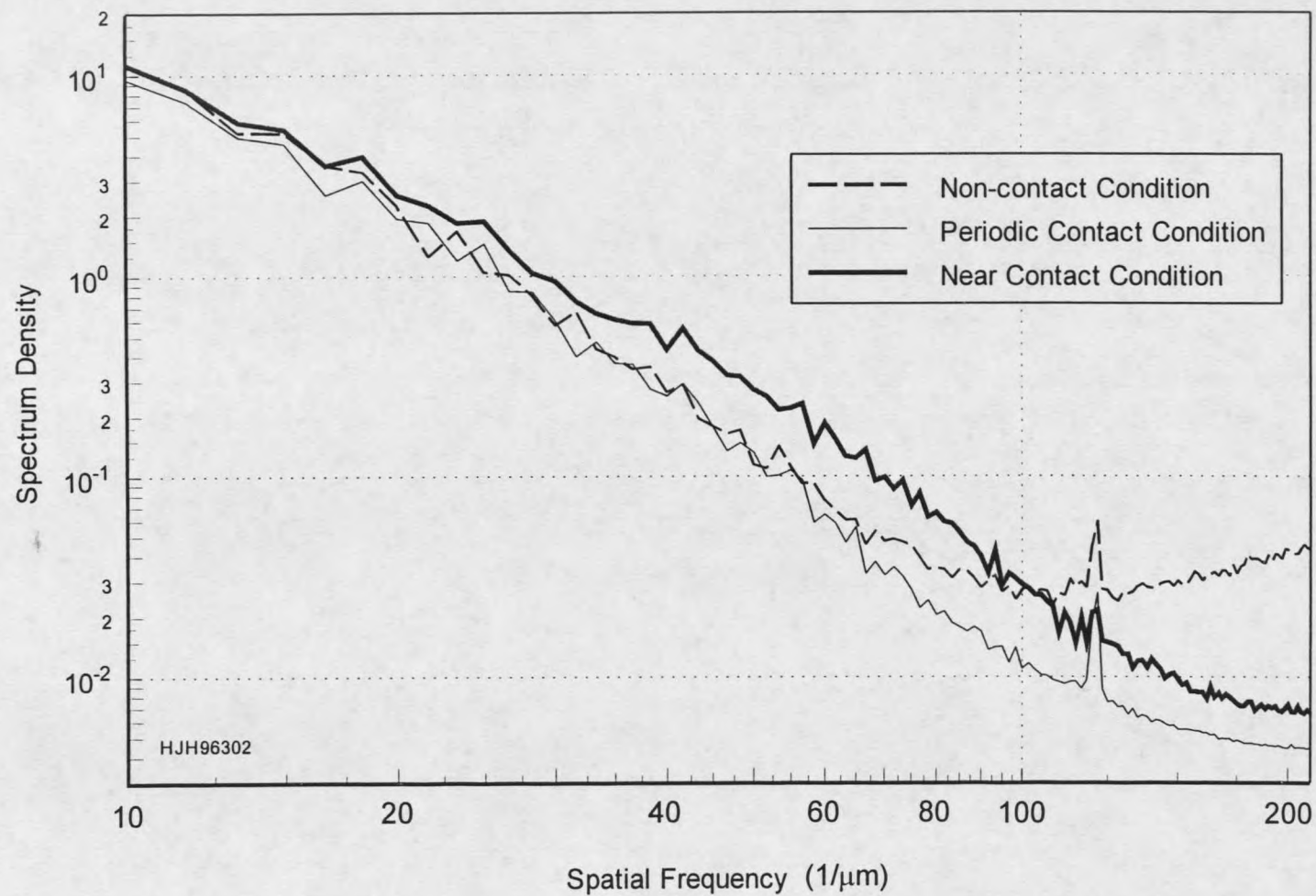


Figure 6.13. Spatial frequency spectrum of the three VC mode operation conditions.

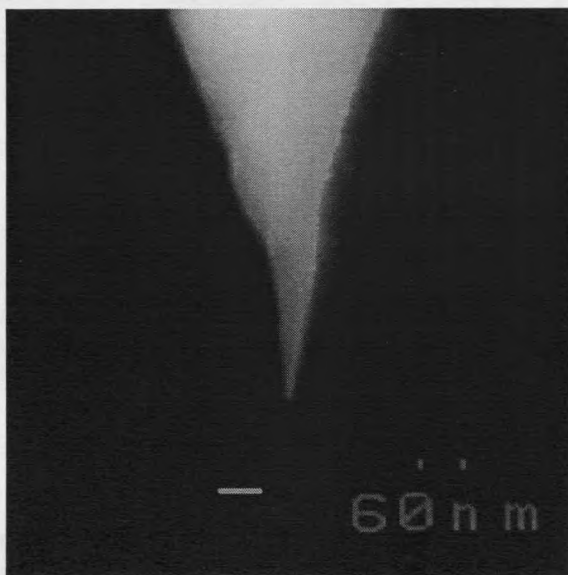
Figure 6.14 shows the SEM images taken before and after those consecutive AFM images. Comparing Figure 6.14(a) with Figure 6.14(b), plastic deformation to the end of the tip can be clearly seen. Since it was proven in another experiment that both the near-contact and non-contact condition do not damage the tip, the change of the tip geometry is most likely to be caused by the periodic contact mode.

Experiment Six: Estimating Lateral Resolution

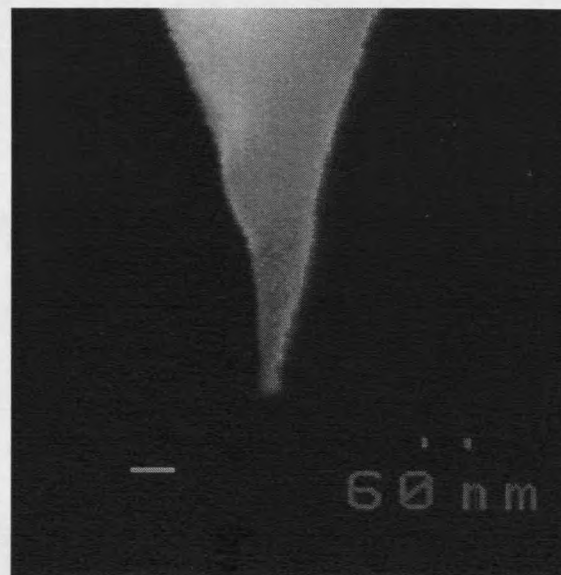
To show that a set of operation parameters is the optimal set, lateral resolution of topographic images must be studied. In order to demonstrate high lateral resolution, a known sample with small surface features is needed. In general, such a sample can be defined to be a standard sample that is used to measure the image resolution.

Figure 6.15 illustrates an AFM image of a niobium film acquired with the near-contact mode. The sample was prepared by e-beam evaporation for characterizing AFM tips.⁸⁶ It includes some known features that are on the order of a few nanometers, which can only be resolved by AFM when operating in optimal conditions. A silicon tip, TopoMetrix model #1650-00, was used for acquiring this image.

To estimate spatial resolution, a small section was cut out from the original image, which is shown in Figure 6.15 (b). Figure 6.16 shows the line profiles of 12 lines extracted from the image of Figure 6.15 (b). In the middle of these line profiles, we can see two peaks. Peak "A" starts to develop near the top, and peak "B" starts to develop near the bottom. On line #16, the peak "B" is only a small spike in one pixel (indicated by a circle). Since that small spike is part of the peak "B", not some random noise, the lateral resolution of this image is equal to its pixel resolution. Since the pixel size is 2 nm, the actual resolution of the near-contact mode is better than 2 nm.



(a)



(b)

Figure 6.14. SEM images of the tip before and after acquiring the three VC mode AFM images.

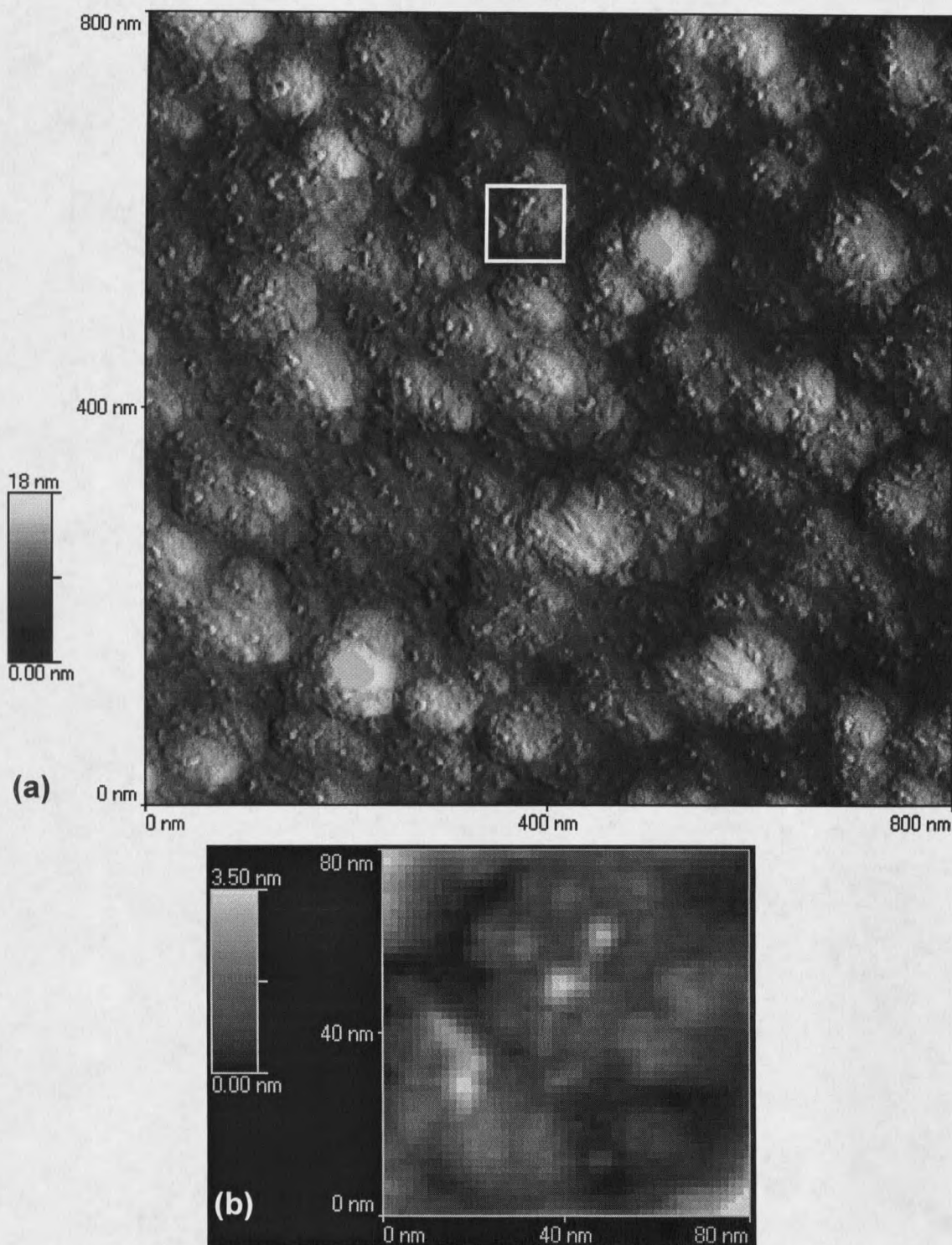


Figure 6.15. Topographic image of a niobium film sample that has the potential of being a standard sample for tip geometry characterization. (a) The image is displayed with pseudo-2D color (90/140); (b) the area of the box in (a), displayed in 1D color.

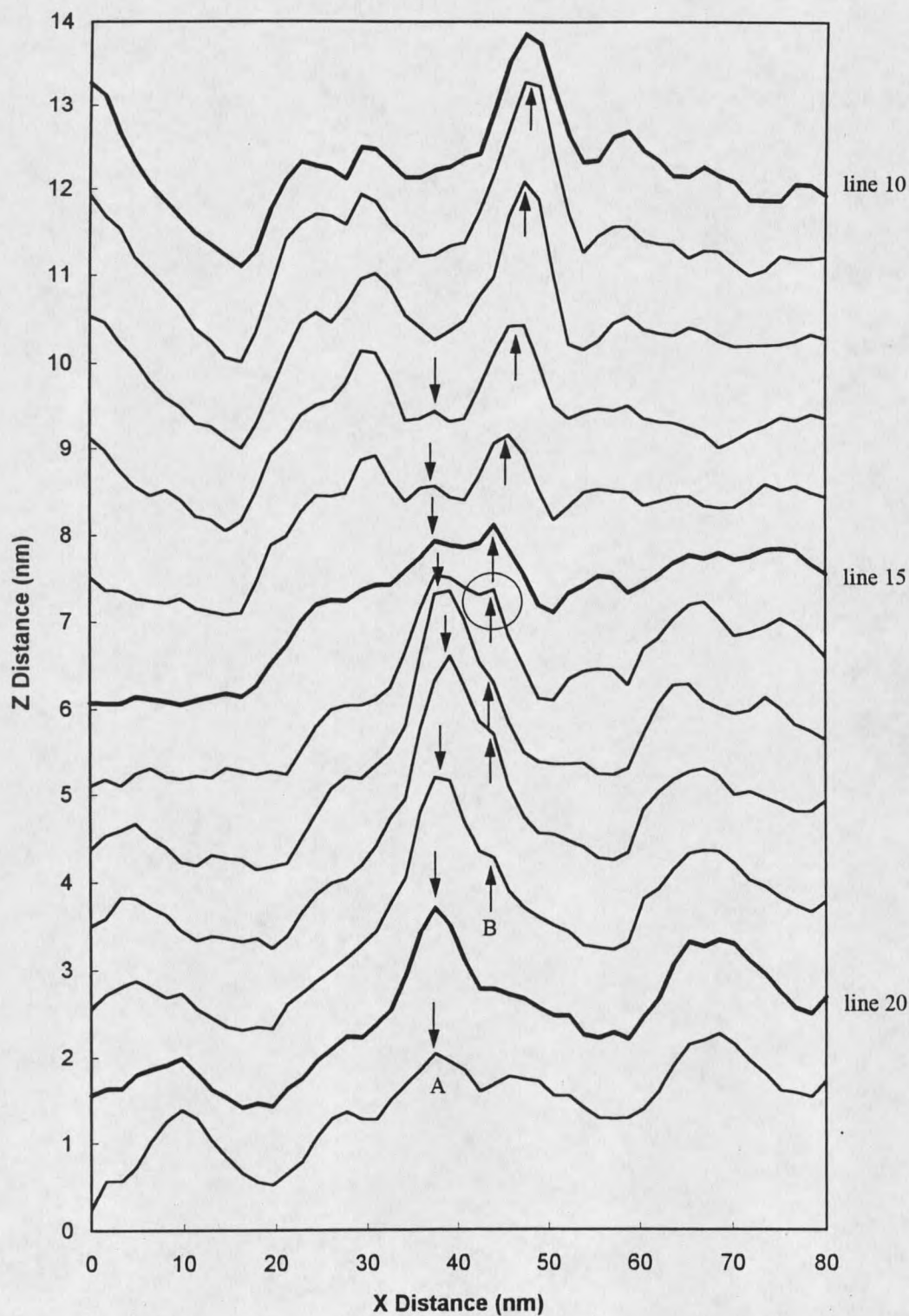


Figure 6.16. A set of consecutive line profile from a zoomed section of the image (Figure 6.15b). The lines are displayed by shifting 1 nm upward from the previous line.

Results and Analysis

Optimal Parameters of VCM Operation

There are several parameters that are involved in the operation of VCM, such as the spring constant of the cantilever, the driving frequency, the cantilever vibration amplitude, the feedback setpoint, and the detection (amplitude or phase) method. The setting of these parameters strongly affects the image quality. The selection of the parameters also depends on the environment. Operating in ambient air is very different from operating in vacuum. The purpose of these studies is to find the optimal operation parameters, which are the ones that can maximize image resolution and minimize tip and sample damage.

Effect of the Spring Constant of Cantilevers. Since the VCM operation relies on the cantilever vibration, the mechanical characteristics of the cantilever are very important. The spring constant is the most important characteristic of a cantilever for VCM operation. To obtain high resolution AFM images, a sharp tip kept close to the surface is required. However, when the tip is brought close to the sample, it is possible that tip-sample contact will damage the tip or the sample. To prevent tip or sample damage, the tip should be maintained in non-contact to the sample. A soft cantilever is readily attracted to contact with the surface by van der Waals force, capillary force, or electrostatic attraction, which requires that it be kept further away from the surface. This large tip-sample gap sacrifices the lateral resolution. Because of its high spring constant, a rigid cantilever can be brought to a few nm (or less) away from the surface without being pulled into contact with the surface (see Chapter 4). Therefore, it can maintain non-contact when operating with a very small vibration amplitude.

There has been a common argument that cantilevers with a high spring constant are not suitable for high resolution imaging because that large spring constant results in poor force sensitivity. For this reason, previous researchers hesitated to use cantilevers with high spring constant for high

resolution imaging. This argument is certainly true in the DC (e.g. continuous contact mode) operation. However, is it still true for vibrating cantilever operation?

In VCM operation, what we detect is not the force, but rather the force derivative. This is because for a vibrating cantilever, the force derivative appears as the change in the effective spring constant of the cantilever. It can be seen in the following equation of motion of the cantilever,

$$m_e \ddot{x} = -k_0 x + F_{ext}, \quad (6.1)$$

where m_e is the effective mass of the cantilever, k_0 is the natural spring constant of the cantilever, x is the displacement of the cantilever, and F_{ext} is the external force. This equation can be written as,

$$m_e \ddot{x} = -k_e x, \quad (6.2)$$

where

$$k_e = k_0 - \frac{dF_{ext}}{dx}, \quad (6.3)$$

is the effective spring constant. What we want to measure is the effective resonant frequency under the external force, that is,

$$\omega_e = \sqrt{\frac{1}{m_e} (k_0 - F'_{ext})}. \quad (6.4)$$

Since the effective resonant frequency is the parameter that affects the feedback signal in VCM, force derivative is the one to determine the feedback that maintains a constant tip-sample gap.

It has been shown that the minimum detectable derivative is governed by the geometry of the cantilever in this way⁶:

$$F'_{min} \propto \sqrt{\frac{k_0}{Q\omega_0}} \propto \sqrt{\frac{\sqrt{k_0}}{Q}}, \quad (6.5)$$

where Q is the quality factor of the cantilever resonance. It was also shown that the major noise, which is the thermal vibration noise, is⁶

$$N \propto \sqrt{\frac{Q}{k_0}}. \quad (6.6)$$

From those results, the increase of spring constant only slightly reduces the sensitivity, on the order of $k_0^{1/4}$. At the same time, it helps suppress the thermal noise on the order of $k_0^{-1/2}$. Therefore, us-

ing a high spring constant cantilever is acceptable.

With silicon cantilevers of high spring constant ($> 50\text{N/m}$), the near-contact mode AFM images with nanometer resolution were obtained on both hard and soft insulators and conductors under ambient conditions. This supports the argument that stiff cantilevers can obtain ultra-high resolution with very small vibration amplitudes.

In vacuum, laser heating reduces the effective spring constant of the cantilever. And static charges increase the attractive force between the tip and the sample, making it difficult to prevent tip-sample contact. More rigid cantilevers are preferred for vacuum operation. With the substantial increase of cantilever's Q factor in vacuum, increasing the spring constant will not cause reduction of sensitivity. Furthermore, high spring constant is needed to suppress the vibration noise that is enhanced by the high Q -factor.

Effect of the Driving Frequency. The VCM operation is based on the relationship between the feedback signal (change of amplitude or phase shift) and tip-sample gap. For a fixed drive amplitude, the amplitude decreases when the tip-sample gap is reduced.⁶ In the phase detection mode, the appropriate phase signal has similar behavior. The value of the set point represents the difference between the final resonant frequency and the driving frequency, which is shown in Figure 6.17. Therefore, for a fixed set point, setting the driving frequency is effectively setting the final tip-sample gap. When the driving frequency is set higher, the final tip-sample gap is larger. If the driving frequency is set too much lower than the natural frequency of the cantilever, the tip will crash into the sample before any feedback can be established.

Usually, when operating in air, the driving frequency is set at the natural resonant frequency when the tip is far away from the sample surface (Figure 6.17(a)). When operating in vacuum, setting the driving frequency at the peak is not possible, because the Q is too high to have a stable peak. A tiny fluctuation can cause dramatic change in the vibration amplitude. To gain stability, the driving frequency is usually set at some value greater than the original resonant frequency.

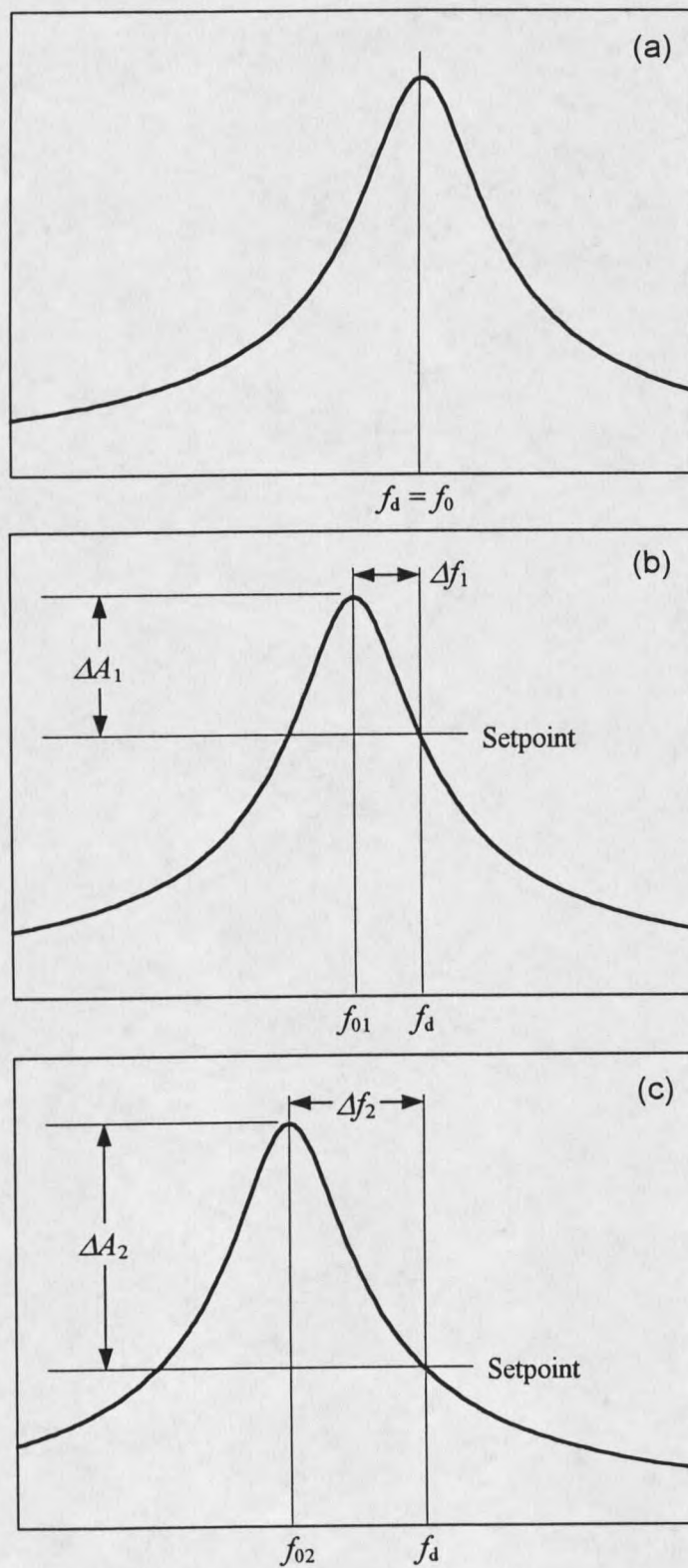


Figure 6.17. Relationship between the drive frequency and the feedback setpoint.

Effect of the Vibration Amplitude. An important parameter that governs the mechanism of tip-sample interaction is the cantilever vibration amplitude. According to equation (4.1), the vibration amplitude, A , can be calculated from: A_e , the oscillation amplitude of the driving voltage applied to the modulating piezo; Q , the quality factor of the cantilever; and p , the piezoelectric gain of the modulating piezo (in distance/voltage).

When the vibration amplitude is too small, the signal-to-noise ratio will be small. The image can be noisy. The noise can further create a false feedback signal that causes the system to be unstable. However, if the amplitude is too large, the tip can gain enough energy to contact the sample surface directly, which could result in significant tip and sample damage. To prevent tip and/or sample damage, the cantilever vibration amplitude should be as small as possible, and be large enough to maintain stable feedback.

Amplitude Detection versus Phase Detection. To detect such a change in resonance characteristics, the commonly used method is slope detection. This detection technique operates with a fixed drive frequency. The change of the cantilever's resonance characteristics is detected by measuring the change of either amplitude or phase in the cantilever vibration. For large amplitude operation, both amplitude detection and phase detection can give good results. However, for small amplitude operation (like the near-contact mode), since the amplitude signal is too small to have a sufficient signal-to-noise ratio, it is better to detect the phase signal.

In VCM operation, the change of phase signal is usually sharper than the change of amplitude when the tip is approaching the surface, which makes the phase detection more sensitive than amplitude detection. The magnitude of the phase signal is generally independent of the driving amplitude, which makes the phase detection method suitable for small amplitude operation. In general, phase detection is the optimal detection technique for VCM operation.

Effect of the Phase Setting. For the phase detection to work properly, the correct phase quad-

rant must be selected. In Chapter 3, Figure 4.16 shows the spectrum of the phase signal of all four quadrants, where only one quadrant can establish proper feedback. In the case of Figure 4.16 (b), the 90° phase is the optimal phase because it has a sharp positive peak. When the resonant frequency shifts to a lower value, this phase signal will decrease. The feedback control electronics has been designed to detect such a decrease. The 270° phase is the opposite phase because it does not fit the feedback condition. Although the 0° phase presents a positive peak, it is not sharp. Compared with the amplitude spectrum, we can see that this phase is off resonance by a significant amount. Although working in this phase can sometimes give an image, the image quality is poor, and tip damage is highly possible.

Working Principle of the VCM Operation

To explain the experimental results and to understand the effect of various operation parameters, we need to study the working principles of VCM. Since non-contact is one of the important objectives of the VCM operation, the concepts of tip-sample contact and non-contact must be clarified. To explain the detail of tip-sample interaction in ambient environment, a microscopic model of the tip-contamination-sample system is developed. Based on this model, the mechanism of feedback establishment of the three important VCM operations is studied. Finally, the mechanism of the possible tip damage in the VCM operations is analyzed.

Concepts of Contact and Non-Contact. When studying VCM operation, one of the important concepts is non-contact between the tip and the sample. When the tip and the sample are significantly far away from each other, they are clearly in a non-contact relationship. However, as they get closer and closer, when will non-contact turn into contact? Since non-contact and contact are a pair of contradicting concepts, we only need to define either one of them, and it is easier to define "contact".

What is contact? How close do two atoms need to be to be called "in contact"? We can define

the concept of contact from the results of the approach process. When the two surfaces are separated, the surface atoms are significantly different from the bulk atoms. When the two surfaces are brought close to each other, at some point the interaction between the closest atoms on the two surfaces is equal to or stronger than the interaction in the bulk. At this point, the difference between the surface atoms and the bulk atoms is minimal. When the tip is pulled away, the original surface atoms will have a hard time "deciding" which side they belong to. Therefore, transfer of atoms between the two materials can be possible. During an approach process, if the interaction at the surfaces is significantly stronger than the bulk interaction, the atoms can be squeezed into free space outside of the contact area, which results in plastic deformation in the interaction region.⁸⁷ In any case, some changes (atom transfer, plastic deformation) might happen to the two pieces of material that are brought close to each other. Such changes are the proof of true "contact." If there is not any change on both surfaces after an approach-separation process, we call this process "non-contact."

Logically, it is possible that the two surfaces come into contact without any atom transfer or plastic deformation, such as, only elastic deformation occurs to the surfaces. However, due to the existence of surface contamination, this is hard to prove in air experimentally. In UHV, however, with a carefully designed setup, one might be able to show this true non-destructive contact. Practically speaking, for any imaging equipment, a non-contact measurement really means a non-destructive measurement. Therefore, using the tip-sample destruction as the guideline to distinguish contact and non-contact is practically appropriate.

Theoretical Model of Tip-Contamination-Sample System. To understand the image mechanism of the VCM, and to develop the optimal operating parameters, it is important to develop a microscopic picture of the tip-sample interaction. In the case of ambient air, there is a surface contamination layer known to exist on the sample surface. Therefore, we need to develop a microscopic model including the tip, the sample, and the surface contamination layers on both the tip and

the sample.

When a vibrating cantilever is moved towards a sample surface, the changes in the vibration amplitude of the cantilever are created by the changes in the force gradient between the tip and the sample. These interactions between the tip and the sample depend dramatically on the environment of the sample. In UHV environment, with an atomically clean tip and sample, the shift of resonant frequency is created by the van der Waals interactions between the probe and surface³⁵. In ambient air, damping of a vibrating cantilever can be created by interaction of the tip with the surface contamination layer.

Any clean surface will be covered with a layer of molecules directly absorbed from their environment that tightly bond to the clean surface. Further contamination that comes to the surface will be loosely bonded to form a liquid-like layer on the surface. Therefore, there is a molecular layer that exists between the liquid-like surface contamination and the solid surface of the sample. This is true for both the sample surface and the tip surface. To damage the tip or the sample, this surface molecular layer must be damaged first. It acts as a buffer layer protecting the surface of tip and sample. Figure 6.18 is the diagram showing the microscopic model of a tip-contamination-sample system.

While the tip is approaching the sample, the force that is applied to the tip can be divided into four ranges. The first is constant force when the tip is sufficiently far away from the sample; the second is attractive force with positive force gradient, where the magnitude of attractive force increases with the decrease of the tip-sample gap; the third is attractive force with negative force gradient, where the magnitude of attractive force decreases with the decrease of the tip-sample gap; the fourth is repulsive force with negative force gradient, where the tip and the sample are in physical contact.

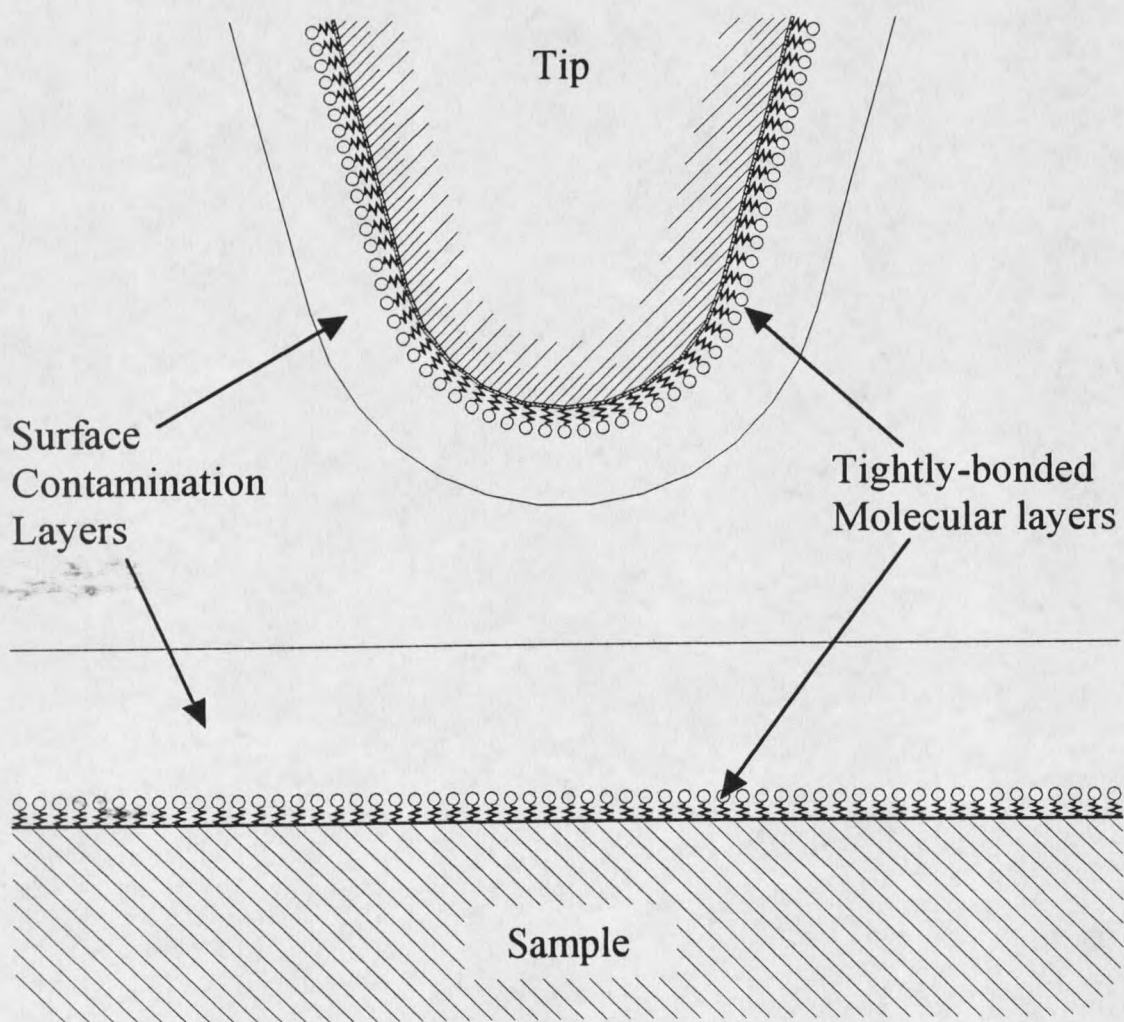


Figure 6.18. Microscopic model of tip-contamination-sample system.

Figure 6.19 shows a molecular model of the tip approach process with the existence of surface contamination layers. A and A' are true non-contact. In this case, the contamination layer on the tip does not touch the contamination layer of the sample and no meniscus is formed between them. The true non-contact situation can be classified into two different stages, A and A'. When the tip is significantly far away from the surface (A), the dependence of interaction force on tip-sample gap is negligible. When the distance between the two surfaces of contamination layers is sufficiently small, the interaction between the surfaces of contamination appears, which is indicated on the force curve (A'). The effect of A' is exaggerated in Figure 6.19, which is too small to measure experimentally.

B is the case when the meniscus starts to form. The tip and the sample are still not in contact with a layer of liquid of contamination between them. The interaction force is attractive with a positive gradient. The change in this force gradient can change the effective resonance frequency of the cantilever, which can establish the topographic feedback in the traditional non-contact mode.

C is the near-contact condition, when the final layer of contamination molecules start to contact. The final layer of contamination molecules is stably bonded to the surfaces. They are a buffer layer between the tip and the sample to prevent direct contact between the tip and the sample. In this case, the force is still attractive, while the force gradient is negative. When the gap between the tip and the sample decreases, the magnitude of the force is getting smaller.

D is the true physical contact condition, when the force between the tip and the sample is repulsive. The molecules of the contamination are forced between the tip and the sample. The interaction between all the atoms in the contact area is no different from the bulk atoms. In this case, atom transfer among the tip, sample, and contamination will occur. This is a major cause of the tip damage.

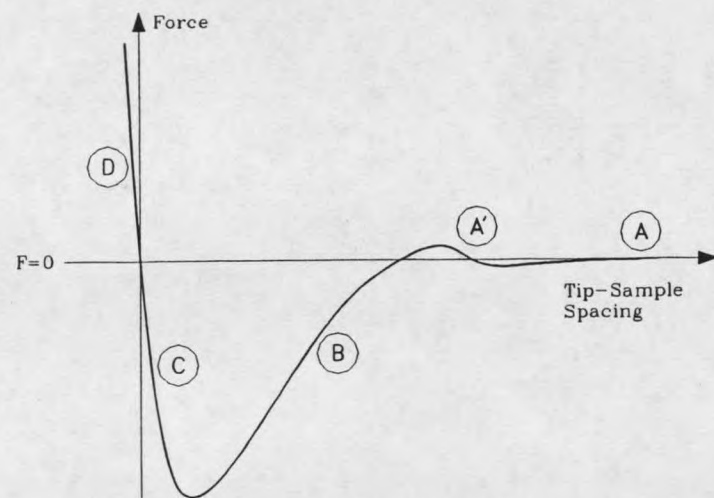
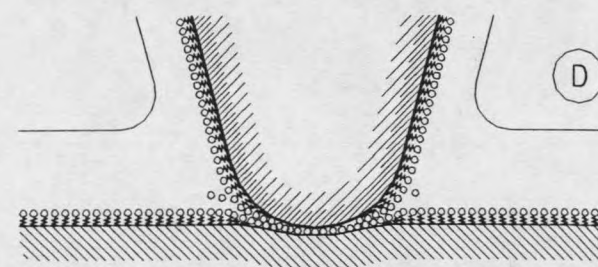
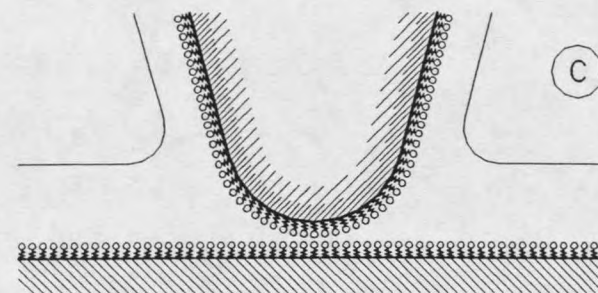
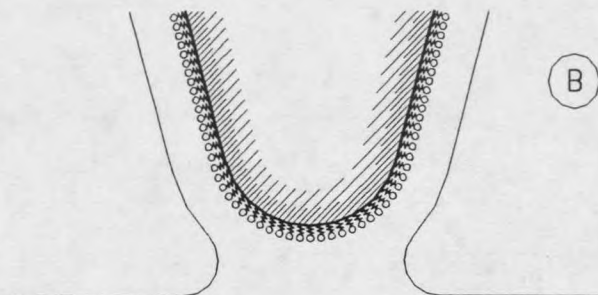
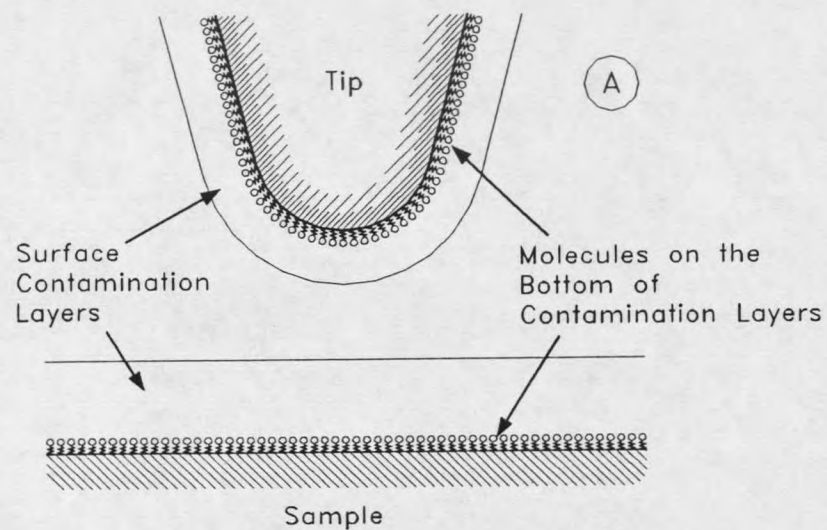


Figure 6.19. Molecular Model of Tip Approaching Process in Ambient Air

Mechanism of Feedback Establishment. For a vibrating cantilever to establish feedback to maintain a constant tip-sample gap, a significant amount of change in the sensor signal should occur when the tip approaches the sample. From the microscopic model, such change is due to the interaction between the tip and the sample surface and/or the surface contamination layers that exist in air. The surface contamination plays a central role in the establishment of feedback for the different VCM operation conditions.

When a vibrating cantilever is brought to the surface, the dominant interaction is initially between the contamination layer on the tip and the contamination layer on the sample. When the two contamination layers make contact, a meniscus forms between the tip and the sample. The capillary force due to the liquid bridge between the tip and the sample causes damping of the cantilever vibration. When the vibration amplitude is small, the capillary force can generate a shift on the resonant frequency of the cantilever that is great enough to allow the establishment of feedback. This is the traditional *non-contact condition*. Under this condition, the tip stays significantly far away from the surface, and physical contact between the tip and the sample does not occur for cantilevers that can balance the capillary attraction. However, the image is strongly influenced by the contamination layer that exists between the tip and the sample. Since the contamination layer is intrinsically liquid-like, the feedback will be unstable, which can cause streaking in the non-contact images.

When the driving amplitude is large, typically greater than 20 nm, the tip will gain a lot of energy. When the tip gets to the top of the contamination layer, the effect of the capillary force is not strong enough to change the vibration characteristics of the cantilever. The change in the sensor signal mostly comes from the interaction between the tip and the sample surface. When significant change in the sensor signal occurs, the tightly bonded molecular layer on surfaces of both the tip and the sample have been destroyed. When the buffer layer is destroyed, the tip can periodically contact the sample directly. This is the *periodic contact condition*.

A new operation condition was first developed during the research for this dissertation. When the vibration amplitude is small (as in the traditional non-contact mode), the cantilever is brought much closer to the sample surface, the tip enters the contamination layer of the sample. At first, the capillary force will pull the tip towards the surface. But if the cantilever has a spring constant greater than the force gradient of the capillary force, the capillary force can be balanced by the spring force of the cantilever. Because the cantilever vibration amplitude is small (< 10 nm), the tip does not have enough energy to damage the tightly-bonded molecular layers on both the tip and the sample, which means that the tip will not contact the surface directly. For this reason, the tip can be maintained at a very small distance from the surface, on the order of 1 nm or less, while maintaining its sharpness. Reduction in tip-sample gap is important for high lateral resolution AFM imaging. This is the *near-contact condition*.

In summary, near-contact condition is the most desirable operation condition for VCM operating in air. It makes use of the contamination, which consist of a molecular layer on clean surfaces and a thin layer of liquid that serves as a lubricant between the tip and the sample. It protects both the tip and the sample to prevent any possible damage due to direct contact. Since the tip can be brought to very close proximity to the surface, ultra-high resolution VCM images can be acquired without any tip or sample damage. Figure 6.20 is the theoretical model showing the relationship between the tip vibration and the contamination layer under the three distinctive conditions.

Mechanism of Tip Damage. A degraded tip can reduce the resolution and produce imaging artifacts. Changes of surface roughness measurements caused by degraded tips have been reported.⁵⁷ With the AFM probes being monitored by SEM with the combined PPM/SEM system, our experiments showed that the tip can be damaged under certain operating conditions. Since the probe tip condition is very important to the quality and repeatability of PPM imaging, it is necessary to analyze the mechanism of tip damage.

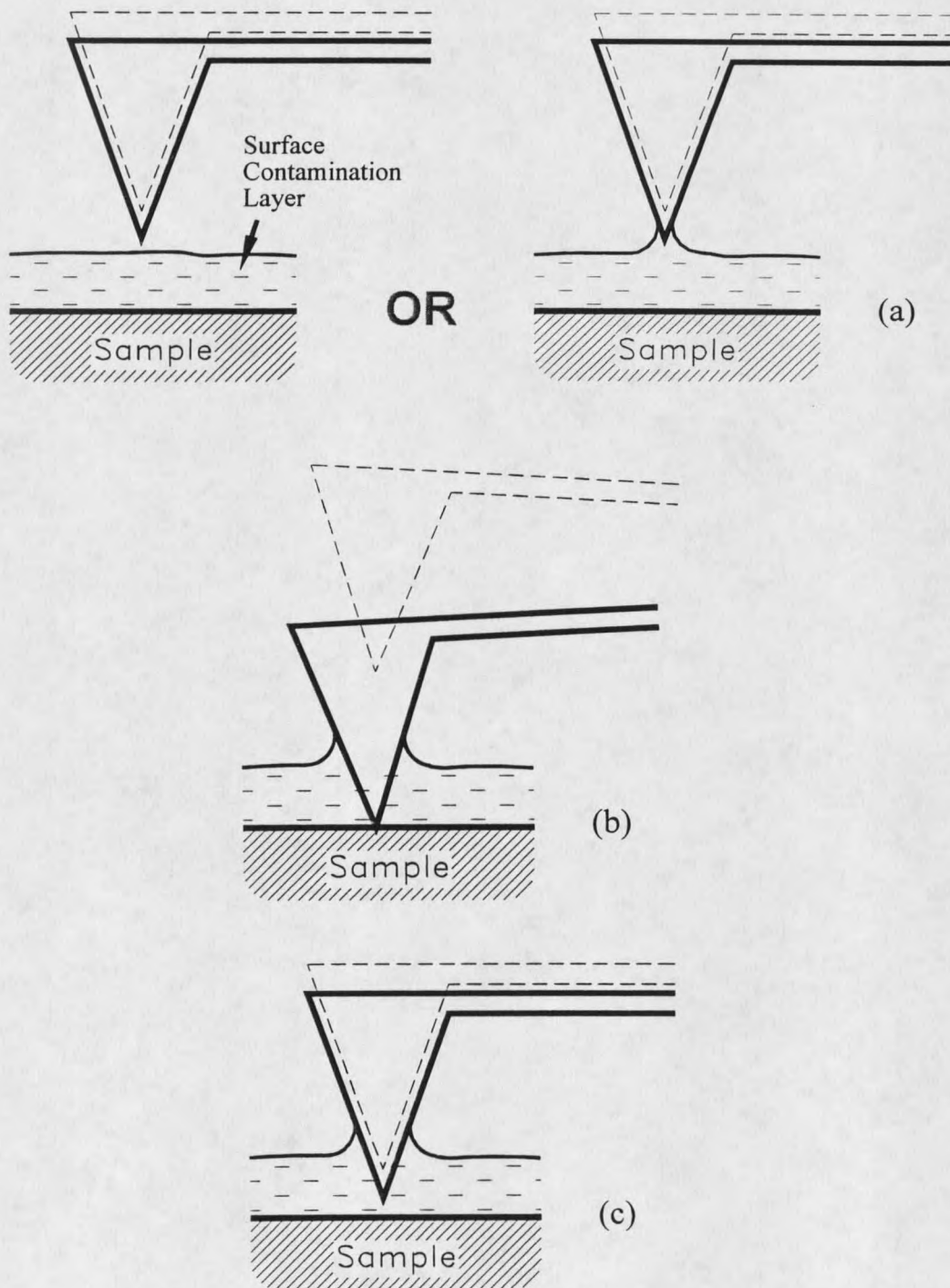


Figure 6.20. Diagrams of the mechanism of the three VC mode operation conditions. (a) traditional non-contact condition; (b) periodic contact condition; (c) near-contact condition.

Several mechanisms of tip damage are possible. First, when the tip strikes the surface with sufficient energy, the molecules in the contamination layer can continue to adhere to the tip. This will increase the diameter of the end of the tip, causing poor lateral resolution. Second, when the tightly bonded molecular layers on both the tip and the sample are destroyed, the tip and the sample come into physical contact, and both the tip and the sample will suffer damage. Third, when the sample is of the same material as the tip, the vacuum welding effect can cause substantial damage in a short amount of time.⁸⁸ Forth, when the tip contacts the sample with sufficient force, the pressure applied to the tip can be high enough to cause plastic deformation of the tip.

According to the microscopic model of tip-contamination-sample system, the layer of molecule bonded to all clean surfaces is an important factor in ambient air environment. This final layer of contamination is difficult to remove, even in a UHV environment. Since most of the tip and/or sample damage is due to the physical contact between the tip and the sample surfaces, this molecular layer can be used as the buffer preventing the damage.

The bonding energy of each molecule in this final molecular layer is on the order of 1 eV. For a cantilever with a spring constant of 50 N/m, the total energy for vibration of 100 nm amplitude is about, $2.5 \times 10^{-13} \text{ J} \cong 1.6 \text{ MeV}$. When the amplitude is 2 nm, the total energy is $1 \times 10^{-16} \text{ J} \cong 0.6 \text{ keV}$. In the periodic contact mode, a common feedback set point is 10% amplitude reduction, which is about 19% energy reduction. Most of this energy loss is absorbed by contamination and the atoms of the tip and the sample near the contact point. Therefore, the energy transfer to the contact region is about $0.19 \times 1.6 \text{ MeV} = 0.3 \text{ MeV}$ per cycle. In the near-contact mode, a common feedback set-point is 70% amplitude reduction, which is about 91% energy reduction. The energy transfer is $0.91 \times 0.6 \text{ keV} = 0.55 \text{ keV}$ per cycle, which is significantly smaller than the periodic contact mode. The energy of chemisorption of an air ion or molecule on a surface is on the order of a few eVs.⁸⁹ For example, chemisorption energy of an oxygen atom on iron surface is about 6 eV.

Table 3. Comparison of energy dissipation.

Operating Mode	Amplitude	Damping	Energy Loss
Periodic Contact	100 nm	10 %	300 keV
Near-contact	2 nm	70 %	0.55 keV
Total chemisorption energy of 10×10 oxygen atoms on iron surface = 0.6 keV			

From the above table, we can see that the energy transfer in near-contact mode is the same order of magnitude as the chemisorpt energy of the final contamination on surfaces. The above example assumes a very small tip-sample interaction area. In reality, considering the size of the tip and contamination on both the tip and the sample surfaces, the number of atoms/molecules involved can be a lot more. Therefore, the near-contact mode generally does not have enough energy to damage the final layer of contamination. In the worst case, the damage can only happen to the molecular layer contamination. Such damage in the molecular layer can be quickly repaired by the liquid-like contamination on the surfaces.

On the other hand, the periodic contact mode has a much larger energy loss to the interaction region. With only 10% damping, the energy loss per cycle is about a thousand times that of the near-contact mode. If the jump-to-contact effect is considered, the amount of the energy transfer is even greater. The large amount of energy between the tip and the sample is the reason for damage to both when operating under such a condition. In addition to our observation of tip damage in the large amplitude periodic contact VCM where the probe strikes the sample on each oscillation, there are other reports of the need to replace probes regularly to maintain the highest quality images⁵⁷, which suggests that the AFM tips are damaged.

Classification of the Three Different VCM Operation Modes

From the loading curve measurement, we can see the interaction force between the tip and the sample. The classification of the three different modes can also be done by studying the interaction

force. On a typical loading curve, there are two important points. The first is the point of touch, where the net force equals zero. At this point, the amount of the repulsive force equals the amount of the attractive force. Since this is the point with minimum potential energy, it is naturally defined as the point of touch. The second is the point of maximum attractive force, where the attractive force reach its maximum.

Figure 6.21 shows an ideal loading curve near the point of touch. The region farthest away from the point of maximum attractive force is the non-contact region where the traditional non-contact mode operates. The region beyond the point of touch is the true contact region where the continuous contact mode and periodic contact mode operate. The near-contact region is only the small region between the maximum attractive force and the point of contact. For tip and sample, their relation is still in "non-contact". The term "near-contact" is used to emphasize that the tip and the sample are very close to each other.

The near-contact region is the most ideal region for topographic imaging, because the tip-sample gap is small enough to achieve high resolution, and the tip and the sample are not in physical contact, maintaining the sharpness of the tip and preventing sample damage. The primary reason that we can work in this region without making tip and sample contact is because of the existence of the surface contamination layers. Operating the VCM in this region requires a stiff cantilever to overcome the capillary attraction, and low feedback setpoint since the damping to the cantilever vibration is very large. To stay in this region requires low energy-dissipation between the tip and the sample, which requires a small vibration amplitude of the cantilever.

One of the advantages of the near-contact mode over the traditional non-contact mode can be seen in the hysteresis of the loading curve. In the near-contact region, hysteresis is minimal. However, when operating in non-contact region, if the feedback loop does not respond fast enough to any high surface feature, the tip will temporarily go into the near-contact region. When the original feedback condition is recovered, the tip will move away to its previous position along the unloading curve. Because hysteresis exists between the loading and unloading process, the feedback loop will operate at a different tip-sample gap. This tip movement results in spike noise in AFM images.

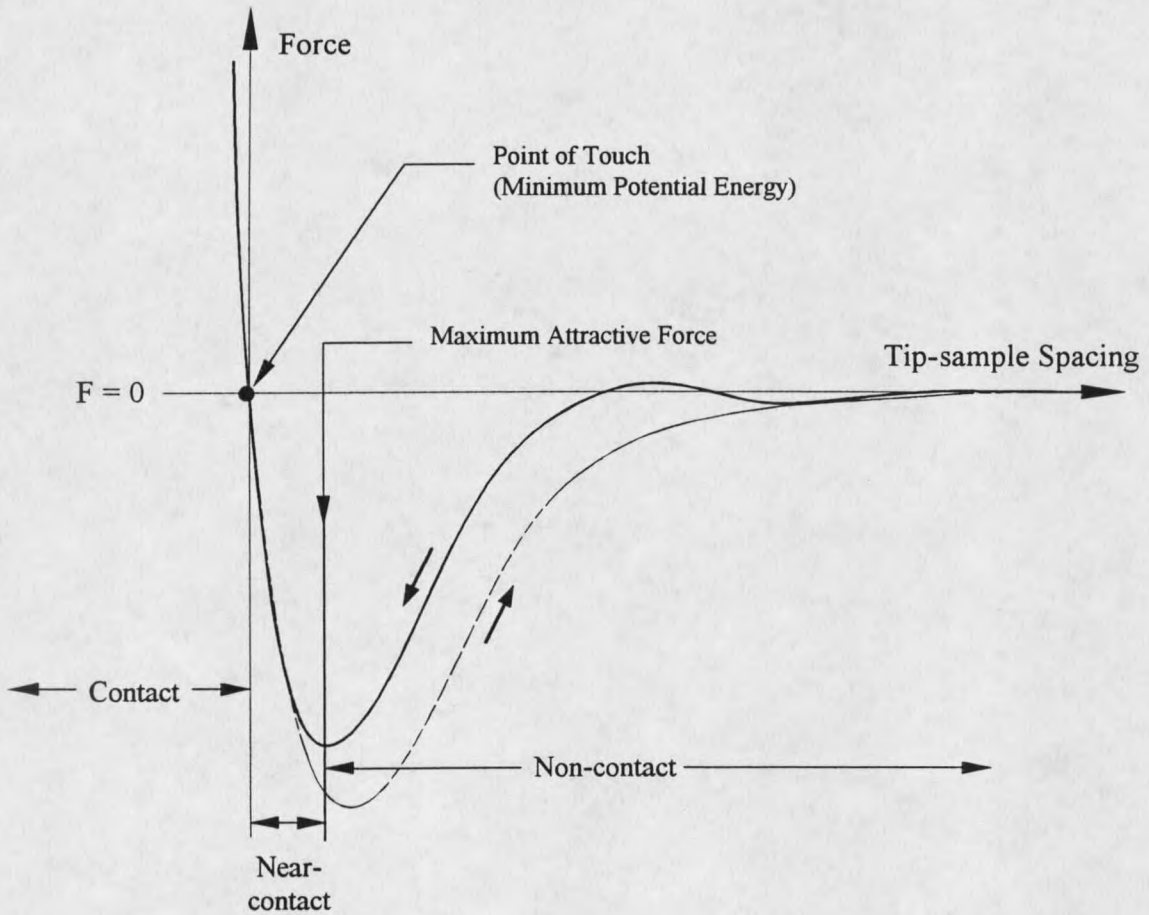


Figure 6.21. Classification of three VC operation modes by the three different stages of tip-sample interaction.

In summary, according to the lowest tip position in the loading curve, the VCM operations can be classified into three distinctive modes, the traditional non-contact mode, periodic contact mode, and the near-contact mode. When a sharp tip (tip radius < 5 nm) is used, the difference in the image quality on the AFM images acquired with these three modes can be clearly seen for an image size of $1\text{ }\mu\text{m}$ or smaller. For images of $10\text{ }\mu\text{m}$ or larger, usually not much overt difference can be seen. The following table lists the comparable items for these three vibrating cantilever operation modes.

Table 4. Characteristics of the three VCM operations.

		Non-contact	Periodic contact	Near-contact
Operation	Amplitude	$< 10\text{ nm}$	$> 20\text{ nm}$	$< 10\text{ nm}$
	Set Point	50% amplitude reduction, or change of phase	10% amplitude reduction	75% change in phase signal
	Feedback Control Parameters	Small	Not sensitive	Large
	Spring Constant of Cantilever	Medium to High	Medium to High	High only
	Detection Method	Amplitude/Phase	Amplitude/Phase	Prefer Phase
Theoretical	Relation to the contamination layer	At the top of the layer	In and out of the layer	Staying in the layer, near the bottom
	Relation to the interaction force	Attractive force, negative force gradient	Repulsive force, positive force gradient	Attractive force, positive force gradient
Performance	Tip damage	No	Yes	No
	Spatial Resolution	Low	High	Higher
	Uncertainty in Z	Large	Large	Small
	Feedback stability	Not stable	Stable	Stable

Examples of Ultra-high Resolution AFM Imaging in Near-Contact Mode

To demonstrate the AFM imaging capability of the new near-contact mode, a number of samples of various different characteristics were imaged. The two most important factors that govern the lateral resolution of the image are: tip-sample gap, the tip radius. To have high lateral resolution, we must have small tip-sample gap and small tip radius. Under the periodic contact condition, although the tip and the sample can be very close (physical contact), a small tip radius cannot be maintained due to the surface damage. Under the traditional non-contact condition, although the sharpness of the tip can be maintained, the tip is too far away from the surface. Under the near-contact condition, the tip can be brought to less than 1 nm from the surface without physical contact between the tip and the sample. Therefore, ultra-high resolution images can be obtained without any tip or sample damage. The following AFM images are shown without filtering.

Metal Film Samples. Metal film samples were imaged in the near-contact mode. Figure 6.15 is an image of a niobium film using a silicon tip. The image was acquired with phase detection in air. After imaging, the tip was inspected by the SEM, and no damage was observed. Figure 6.22 is an image of an aluminum-coating surface. Since this is a rough surface, a high aspect ratio VLS silicon tip was used. The image was acquired with amplitude detection in air.

Semiconductor Samples. Figure 6.23 is an image of cleavage surface of the GaAs wafer sample. The surface has been passivated by $(\text{NH}_4)_2\text{S}$. The image was acquired in vacuum with a VLS silicon tip. The topographic image (Figure 6.23(a)) clearly shows three atomic steps on the surface. These three steps can also be identified by the three peaks in the topographic histogram (Figure 6.23(b)).

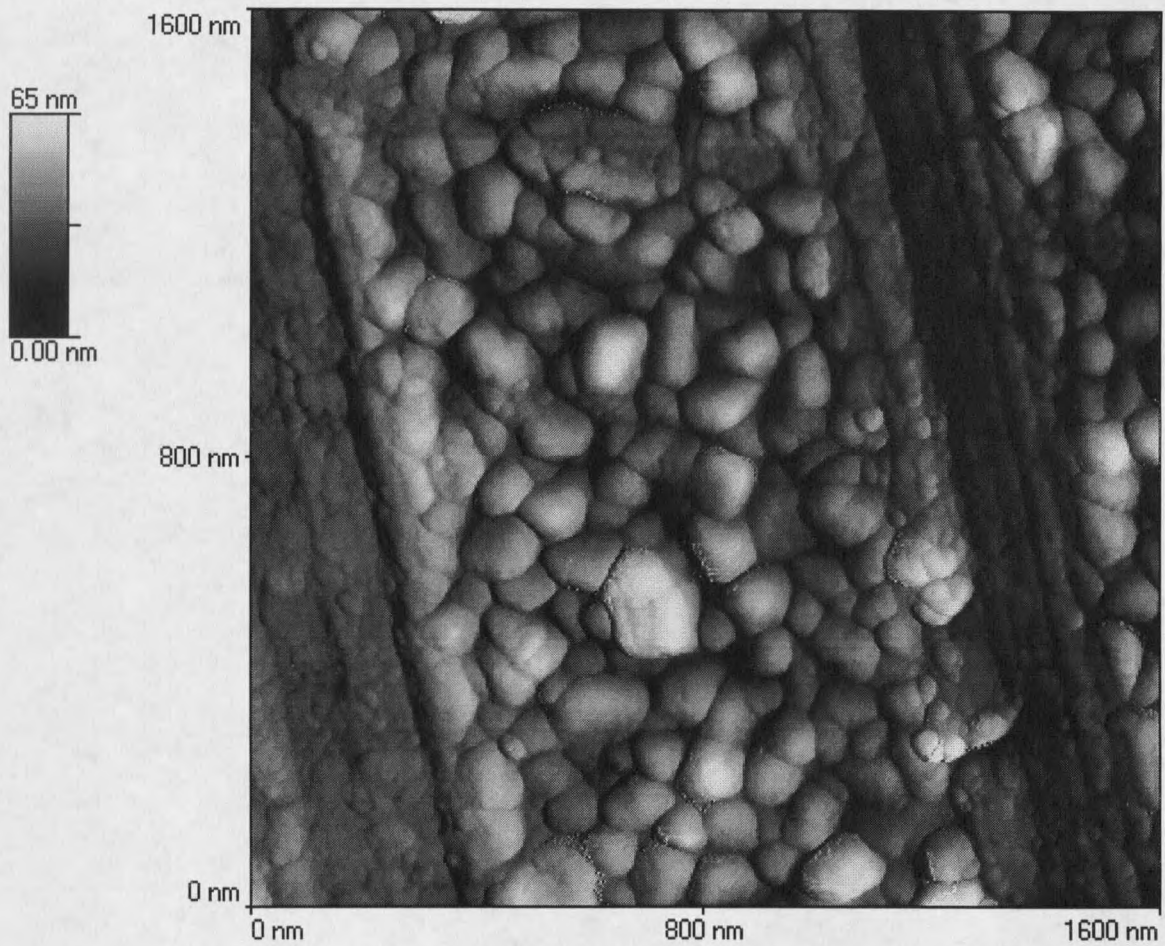


Figure 6.22. Aluminum film on a silicon substrate

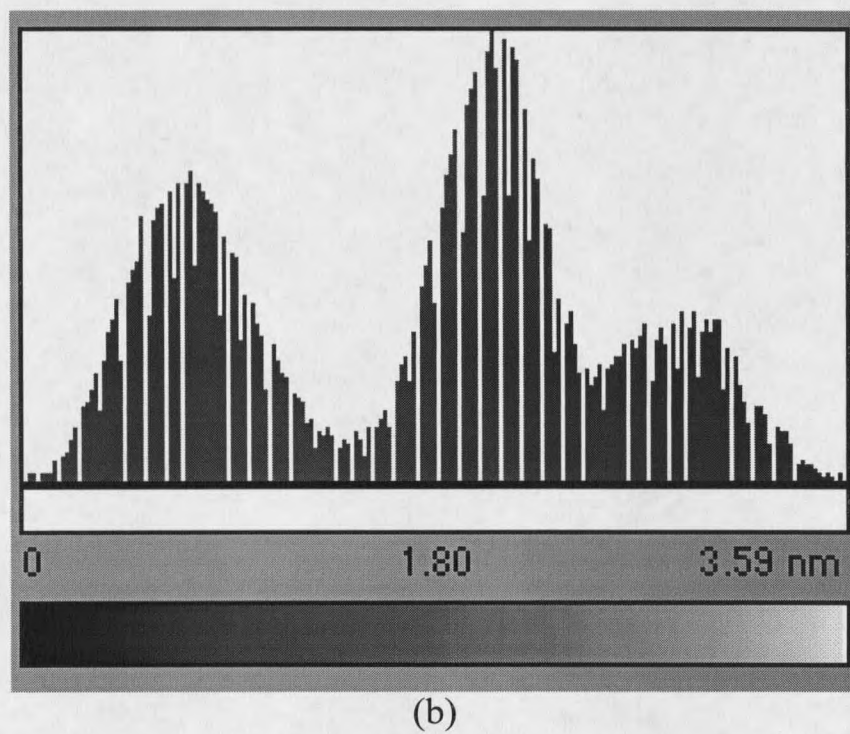
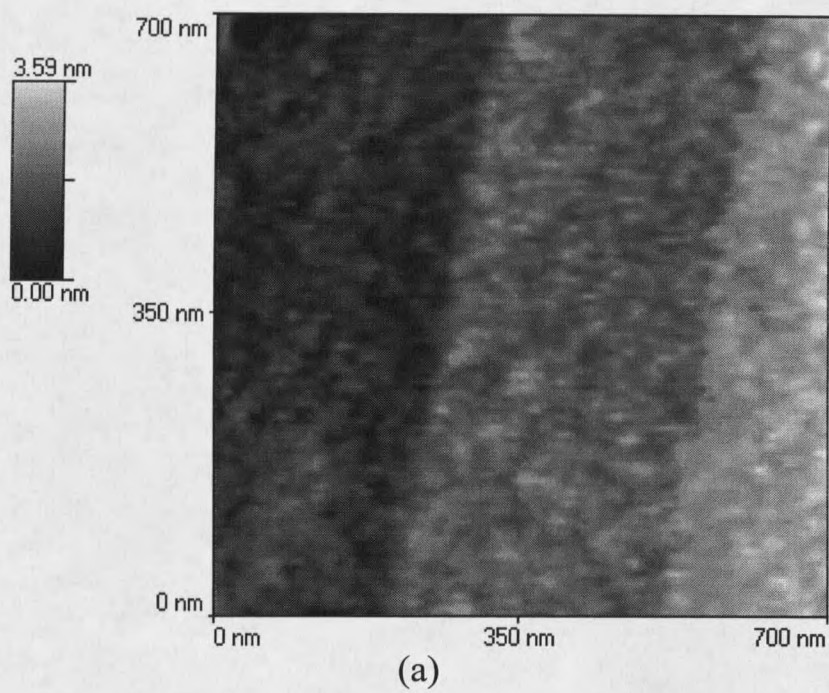


Figure 6.23. Atomically flat cleavage surface of GaAs. (a) Topographic image; (b) Histogram of the topographic data.

Insulating Samples. Insulating samples can present serious problems due to the static charge built up on the surface. For SEM, a thin layer of conductive (metal or graphite) coating is usually applied to the surface to remove the surface charging. However, the surface coating usually covers nanometer-scale features on surfaces. For AFM, static charge can result in JTC in the VCM operation. However, if a rigid cantilever is used, high-resolution images can be obtained in the near-contact mode without any surface treatment. Figure 6.24 is an image of polished alumina-silicate glass substrate, used for a computer hard disk, acquired in air. A silicon cantilever with a spring constant of 50 N/m was used.

Soft Polymer Samples. For soft polyimide and polymer film, contacting the sample can cause substantial sample damage. With near-contact VCM, high-resolution images can be repeatedly obtained without any sample damage. Figure 6.25 is an image of an epoxy acrylic coating.⁹⁰ The worm-shape features (indicated by the arrows) are the wax that is mixed in the coating. Figure 6.26 shows two kinds of polymer films with different surface textures that show a lateral resolution of better than 2 nm.

Biological Samples. Like polymer samples, biological samples usually have a soft surface. Contacting the sample can cause surface damage and deformation. Figure 6.27 shows the surface of a piece of hair of a 3-month-old human infant girl. Since there was oil on the surface, the capillary force between the tip and the sample was stronger. The tip was occasionally pulled to contact the sample, which can be seen from the small spikes in the AFM image. Cantilevers with a higher spring constant are needed to maintain stable feedback for this sample.

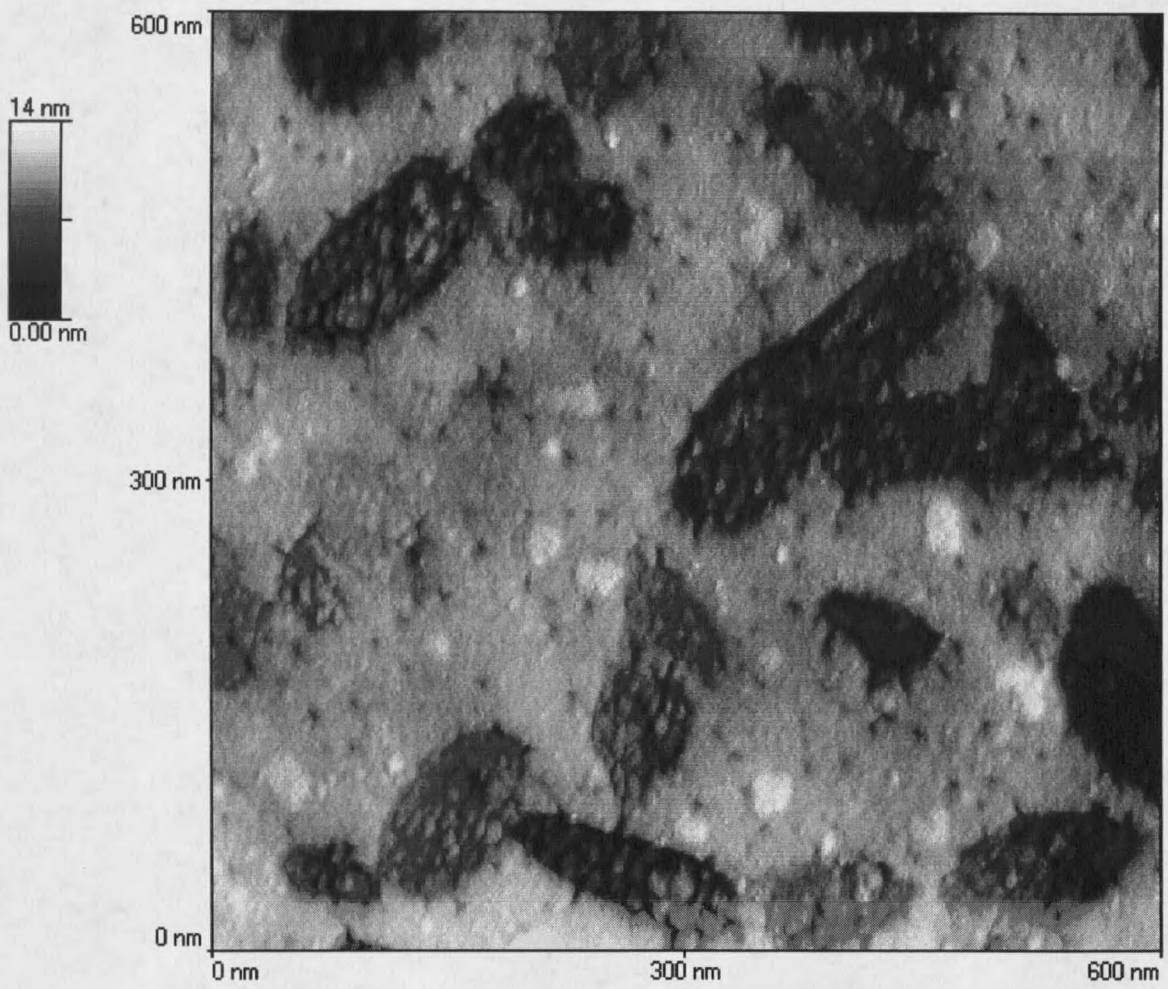


Figure 6.24. Polished alumina-silicate glass substrate.

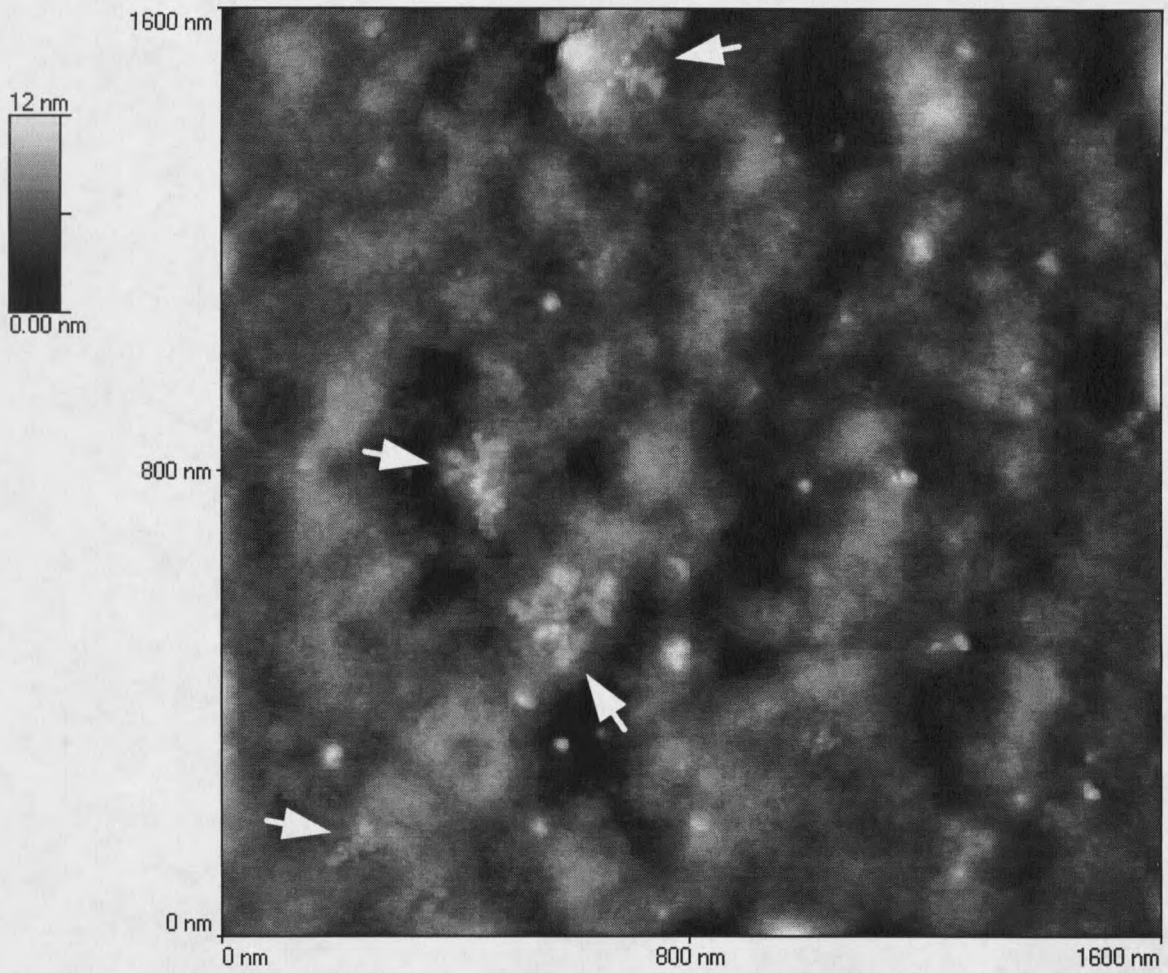


Figure 6.25. Epoxy acrylic coating.

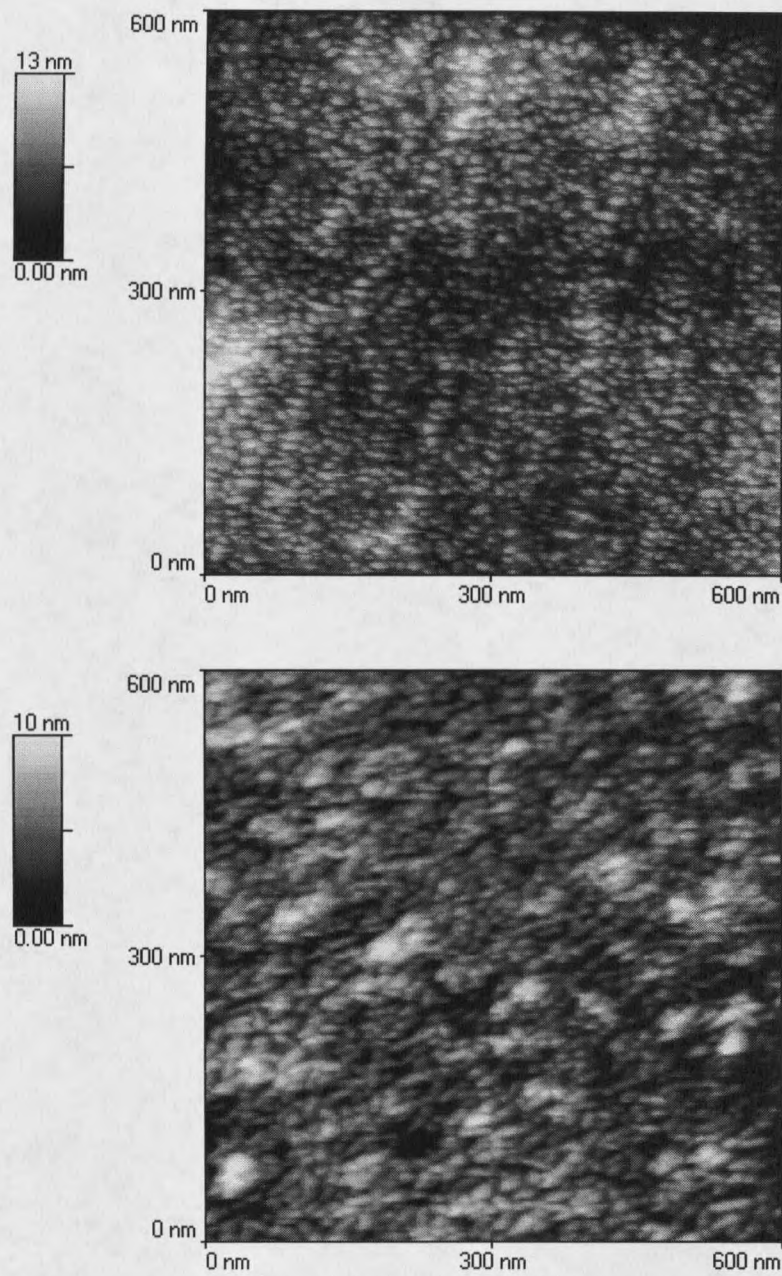


Figure 6.26. Two types of soft polymer thin-film samples.

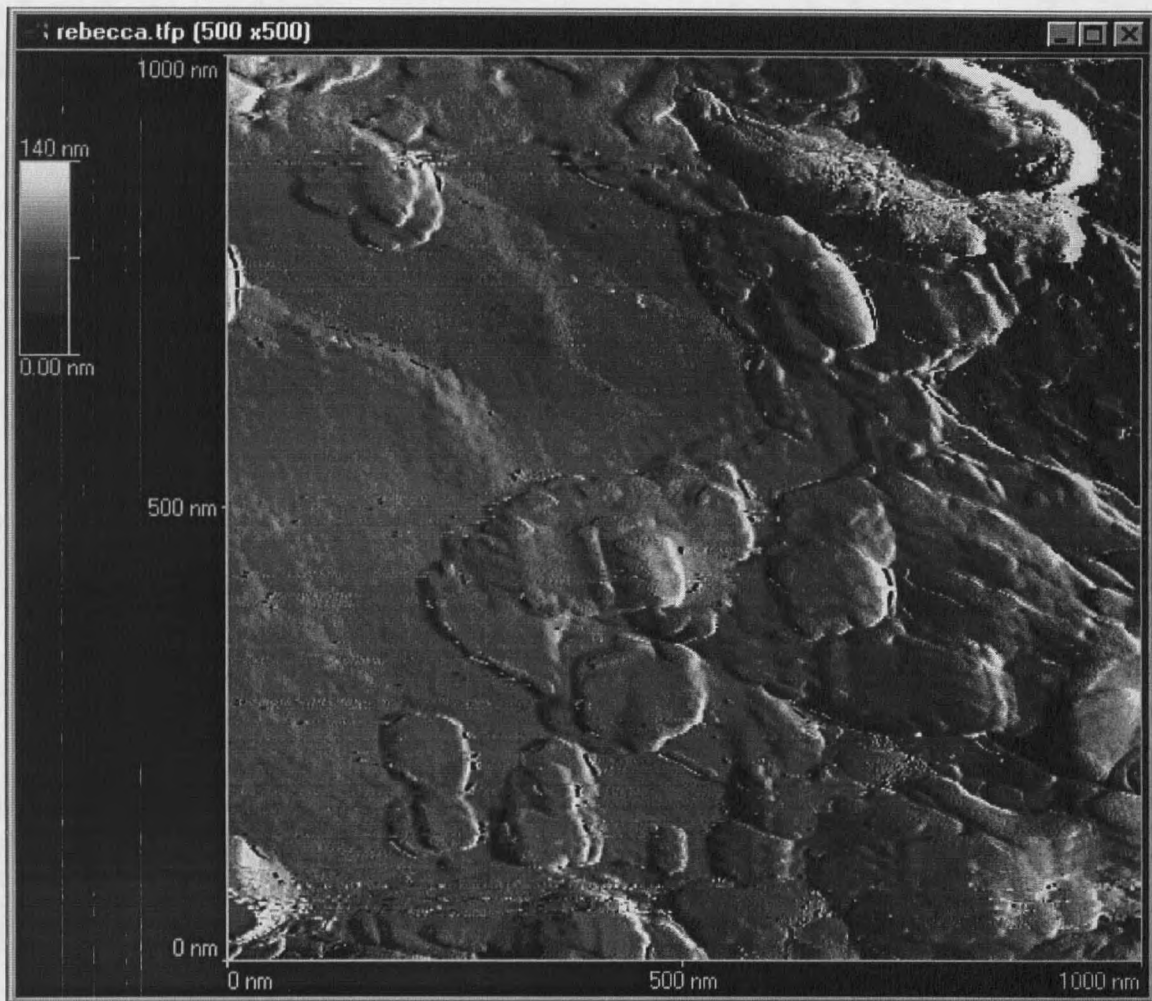


Figure 6.27. Human infant hair sample.

Conclusion

Since the vibrating cantilever mode (VCM) was proposed in the first AFM paper, two major operation modes were developed: non-contact mode and "tapping" mode. When operating in ambient air, these old methods present a number of problems. In the traditional non-contact mode, the existence of surface contamination layer results in large tip-sample gap, which limits the lateral resolution of the microscopy. When the tip is brought into the contamination layer, the capillary force between the tip and the sample can capture the tip to the surface, which stops the vibration of the cantilever. In the "tapping" mode proposed later, the cantilever is set to vibrate at much larger amplitude (> 20 nm) which results in periodic contact between the tip and the sample. Such physical contact, although it can be very light, results in tip and/or sample damage when operating in air, which also limits the lateral resolution.

To understand the tip-sample interaction and the working mechanism of VCM, we performed a number of experiments. With the combined AFM/SEM instrument, we observed three types of nanometer scale tip damage: contamination build-up (Figure 6.2c), wear-off (Figure 6.5b), and deformation (Figure 6.14b). For the first time, we observed the sample damage as the result of minimal periodic contact when the sample is scanned in ambient air. The experiment was performed on a thin polyimide coating on glass.

We discovered a new region at close proximity of the surfaces under ambient air condition called the near-contact region. The existence of this new region can be clearly shown by observing the feedback sensor signal while reducing the tip-sample gap. When the tip is imaging the sample in this region, the tip-sample gap is minimized and physical contact between tip and sample does not occur. Both of these factors result in maximizing the lateral resolution of AFM. From the theoretical point of view, this region can be described as attractive force region with negative force gradient (that is, the magnitude of the attractive force decreases when the tip-sample gap reduces).

We developed a new theoretical model of the tip-contamination-sample system under ambient

air to explain the near-contact region and the working mechanism of the VCM. The surface contamination layer consists of a molecular layer that has stable bonding to the clean sample surface and a liquid layer of a few nanometers thick above the tightly-bonded layer. When the tip is in the near-contact region, the repulsive force is due to the direct interaction between the tightly bonded molecular layers on the surface of the tip and the sample. Since a certain amount of energy is required to destroy this molecular layer, resulting in direct contact between the tip and the sample, cantilevers with small vibration amplitudes can preserve the molecular layers and further prevent tip and/or sample damage due to direct physical contact. Although this structural model of surface contamination is supported by all of our observations, direct experimental proof is still needed.

Based on the understanding of tip-sample interaction and the working mechanism of VCM, we developed a new operation mode for the VCM, which we call the near-contact mode. The three major aspects of the near-contact mode are large spring constant, small vibration amplitude, and low feedback setpoint. The large spring constant of the cantilevers is to prevent JTC when the tip enters the contamination layer on the sample surface. The small amplitude is to ensure minimal energy transfer between the tip and the sample and to preserve the final layer of the contamination molecules, and to further maintain the sharpness of the tip. The low feedback setpoint is used to bring the tip into the near-contact region. With a cantilever that can be controlled when the tip is submerged in the surface contamination layer, using a small vibration amplitude and a low setpoint, the tip is maintained in the near-contact region and the lateral resolution is maximized. We have shown that the operation of near-contact mode is very repeatable in getting stable, high-resolution topographic images in air. The success of this new imaging mode was demonstrated on various types of samples, which shows that high quality VCM topographic images can be obtained without the use of large vibration amplitude.

In operations with large vibration amplitudes, the tip-sample relation is periodic contact, where the tip strikes the sample at the bottom of each oscillation. The large energy transfer between

the tip and the sample destroy the final molecular layer on the surfaces of them, which results in direct physical contact of the tip and the sample. With this operation mode, the topographic image is stable (and appears to have low-noise). However, the tip is damaged, and the detail information of the sample topography is lost. In a comparative study on the three different VCM operation modes with a hard insulating sample, the near-contact mode performed the best and resulted in a lateral resolution that was significantly higher than the traditional non-contact mode and periodic contact mode.

CHAPTER 7

IMAGING SURFACE ELECTRICAL POTENTIAL

Introduction

Surface electrical potential is an important physical property of many surfaces. It depends on a variety of material parameters such as the work function, localized space charge, and surface layers. Imaging the surface potential with a high spatial resolution can allow us to study the localized electrical property of the sample. For example, on a semiconductor sample, we can study the local variation of the dopant concentration. And on a ferroelectric sample, localized electrical charges and domain structure can be imaged.

In this chapter, we first describe the working principles of the scanning Kelvin-probe force microscope (KFM), used to measure surface electrical potential. Then we use a piezoelectric sample to show that KFM removes electrostatic artifacts from topographic images and creates potential images. The time dependence studies on this sample also reveal the electrical charges in the surface contamination layer. To test this potential measurement technique, we designed a standard sample, which can provide a known localized potential difference. Finally, to illustrate a practical application of the KFM, dopant concentrations were measured from surface potential imaged on the surface of a cross-section of a GaAs multilayer sample.

Scanning Kelvin-probe Force Microscopy

A traditional method of measuring the surface potential difference between two materials is

the Kelvin method, or the vibrating capacitor method. In this method, two surfaces are placed very close to each other, with one material vibrating against the other. Due to the change of the tip-sample gap, there is a change in the capacitance. When there is a potential difference between the two materials, the change of capacitance results in a change in the current that can be detected. To measure the potential difference, an additional balancing voltage is applied between the two materials. When the current vanishes, the applied voltage is a measurement of the potential difference.

By applying the concept of the Kelvin method to a vibrating cantilever AFM, a scanning Kelvin probe force microscope is developed.³⁹ In this method, a metalized AFM probe (cantilever-tip assembly) is used to detect the surface potential. The tip is kept very close to the sample surface with the VCM topographic feedback. To detect the surface potential, instead of vibrating the cantilever mechanically, an oscillating voltage is applied to the cantilever with a frequency close to the resonant frequency of the cantilever. When there is a potential difference between the tip and the sample, the cantilever will have detectable vibrations. An external DC bias voltage is added to the cantilever, to give a measure of the potential difference between the metal tip and the sample, when cantilever vibration vanishes.

Figure 7.1 is a diagram that illustrates the working principle of the KFM. The surface potential changes or localized static charges will contribute to the tip-sample interaction. When the surface electric field is not canceled by any external bias, these localized electrical structures will show up in a regular VCM topographic image as electrostatic artifacts. These electrostatic artifacts actually contain information about the electrical properties of the surface. When an additional balancing voltage is applied to give a quantitative measurement of the surface potential change, it also assists the elimination of those electrostatic artifacts from the topographic image. An example of removing the electrostatic artifacts in a topographic image and obtaining surface potential will be shown in the next section.

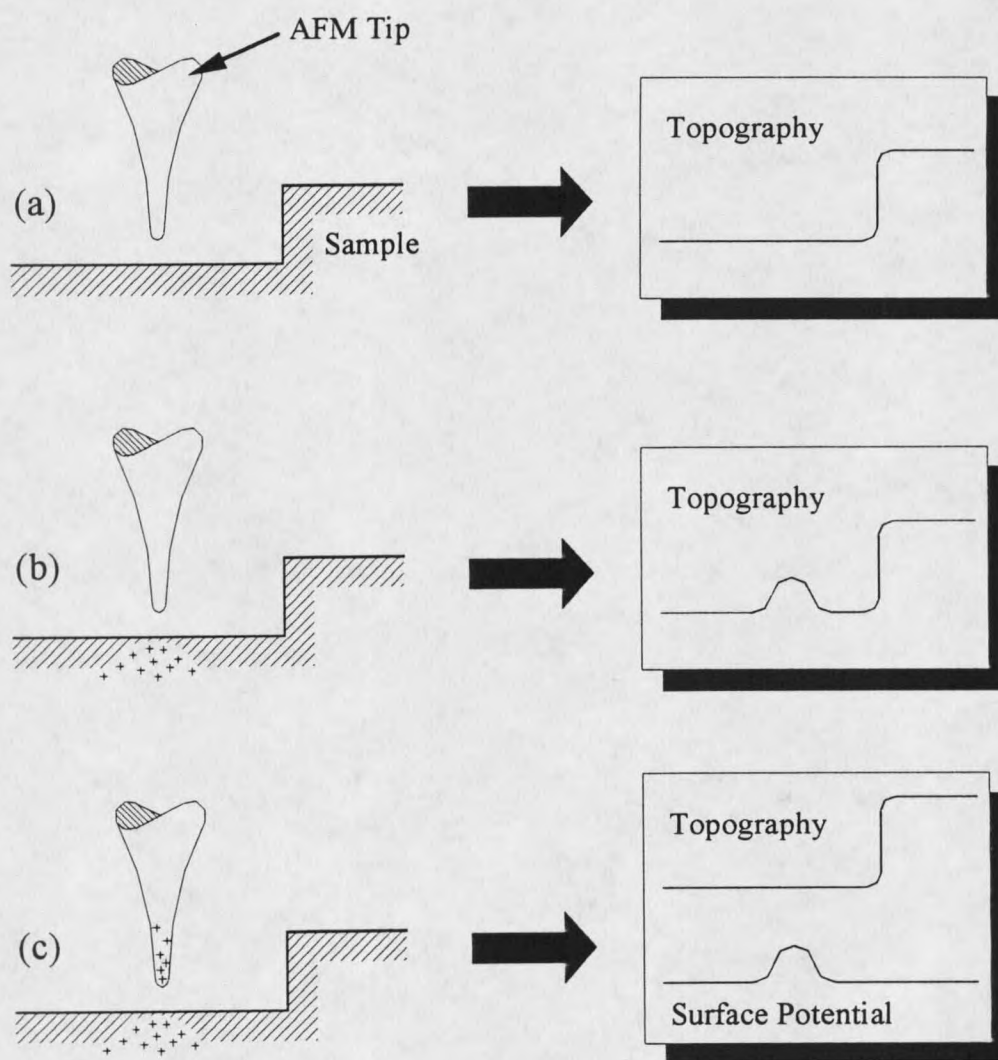


Figure 7.1. Separating the surface electrostatic information from the topographic information. (a) There is only the non-contact topographic feedback loop. The sample has no surface potential change. (b) There is localized surface charge on the sample. (c) Topographic feedback plus the potential feedback for the sample with localized surface charge.

Imaging the Surface of a PZT Film

Piezoelectric materials, like PZT, have highly polarized surfaces. When in ambient air, their surface contamination can contain significant amount of charges that are trying to neutralize the surface electrical field. With KFM, these surface charges may be observed.

Figure 7.2 shows an example that demonstrates the working principle of KFM on a sample of PZT ferroelectric ceramic film.⁹¹ Figure 7.2(a) shows the topography image with no additional bias voltage applied to the AFM cantilever. To enhance the visual effect of the surface features, the derivative image of the false-color height image, Figure 7.2(a-1), is shown as Figure 7.2(a-2). In this pair of images, an obvious feature is a bright line running across the field of view (indicated by the arrow). After this image was measured, the surface potential feedback loop was turned on, which applied a balancing DC bias voltage to the cantilever to null the field between the tip and the sample. Figure 7.2(b) shows the topographic image with the additional bias voltage. Figure 7.2(c) shows the signal of the bias voltage. From Figure 7.2(b), the bright line nearly disappears, leaving a small trench. On the other hand, a bright line shows up in the surface potential image, Figure 7.2(c). This line is a scratch mark on the surface of the ceramic film. Since the material is ferroelectric, the surface damage left a significant amount of residual charge, which contributed to the local surface potential change.

To further illustrate the effect of the additional surface potential feedback loop, a line profile across the scratch mark is extracted from the images shown in Figure 7.2(a) and (b). Figure 7.3 shows these two line profiles. The difference between the two line profiles is dramatic. Two observations can be concluded from the line profile. The first is that the high peak, which is the bright line in the images, is completely removed from the image by the surface potential feedback loop. The second is that the fluctuation on the surface topography is reduced significantly when the surface potential feedback loop is on. This effect can also be seen by comparing Figure 7.2(a-2) and (b-2). The instability on the topographic image is due to the surface contamination layer. Because this layer is not electrically neutral, it adds electrical artifacts to the topographic image.

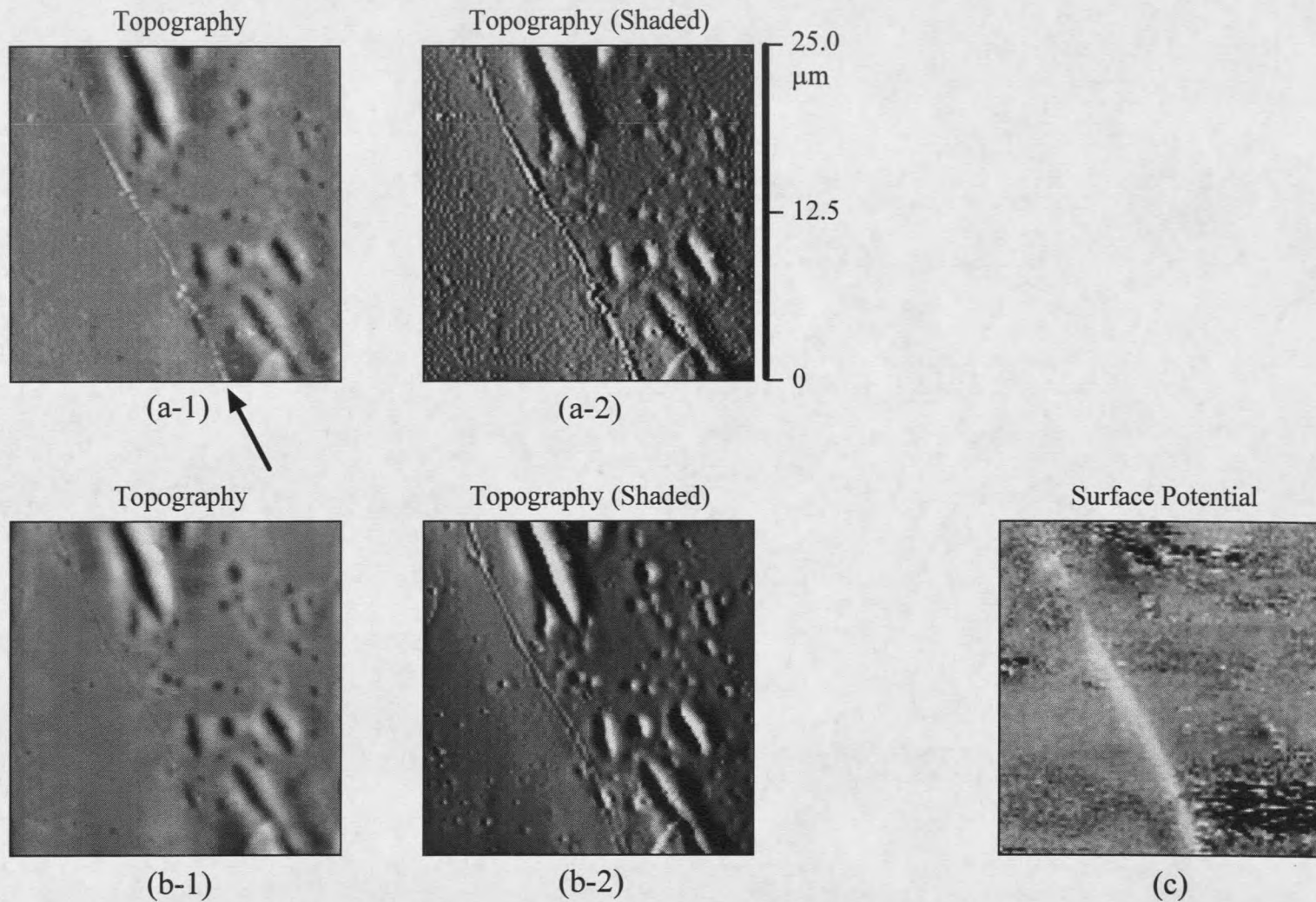


Figure 7.2. Images of a PZT ceramics surface: (a) before, (b and c) after the surface potential feedback loop is turned on.

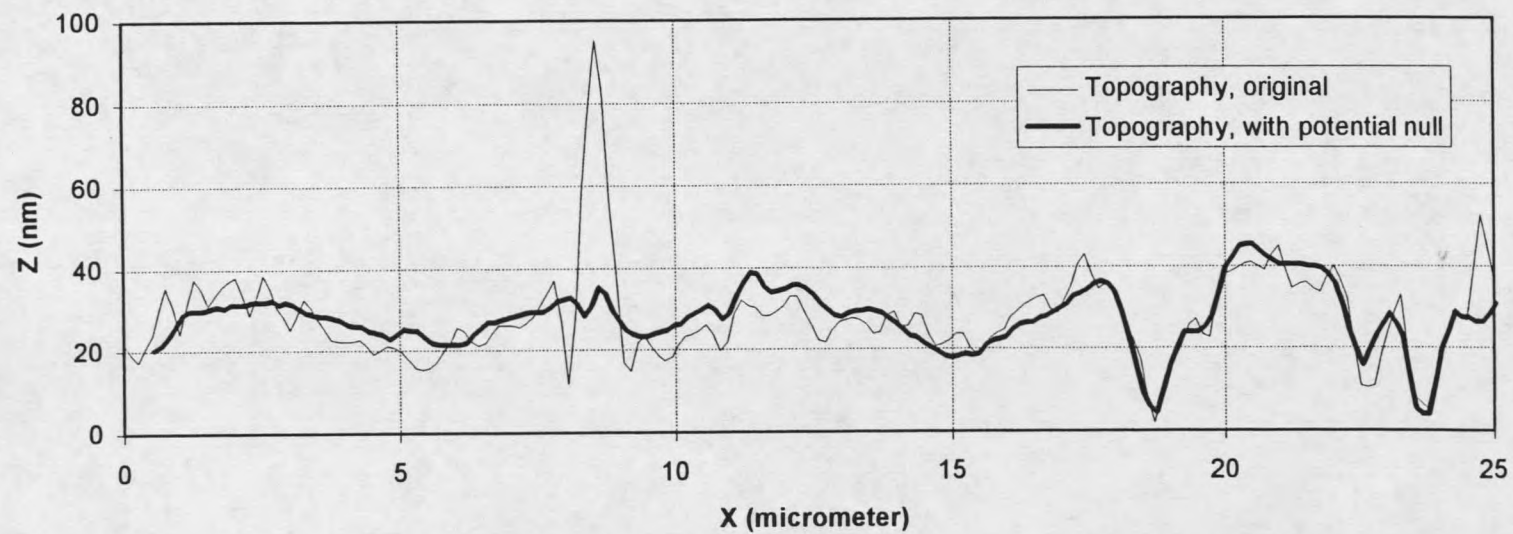


Figure 7.3. Comparison of topography line profiles before and after the surface potential feedback loop is turned on.

Figure 7.4 shows a time dependence experiment, where a set of topographic and potential images were acquired on the same sample surface area ten minutes apart. The topographic images, Figure 7.4 (a) and (b), are very much the same. However, the potential images, Figure 7.4 (c) and (d), have significant differences. The only repeatable feature is the bright line, which is the result of the charge in the scratch. The other “noisy” features are surface charges that are not stable. Since the PZT ceramic is an insulator, the charge distribution can not vary in such a short time, therefore these fluctuations in the KFM images are due to the charges in the surface contamination layer.

Designing and Studying a Standard Sample

For evaluating the performance of a surface potential imaging system, a standard sample was developed (Figure 7.5). It consisted of two sets of “fingers” of chrome film deposit on top of an undoped silicon wafer, so that voltages can be applied to either side of the “fingers” separately, to generate controllable surface electrical potential. Four groups of “fingers” (A, B, C, and D) with different width are built on each sample.

Figure 7.6 shows a pair of images of this sample acquired on a TopoMetrix Explorer™ system. During the measurement, the set of electrodes labeled with the letter “A” in Figure 7.6 were always grounded. The other set of electrodes, labeled “B”, was applied with a voltage controlled by the software. The resonant frequency of the cantilever was 73.2 kHz, and the Q-factor 360. The images were acquired with the mechanical vibration amplitude of 11 nm and an AC bias amplitude of 10 V. The operational frequency for the second feedback loop for potential measurement was set at 86.6 kHz.

Sample: PZT film
Image Size: 25 μm
Resolution: 100 $\times$ 100

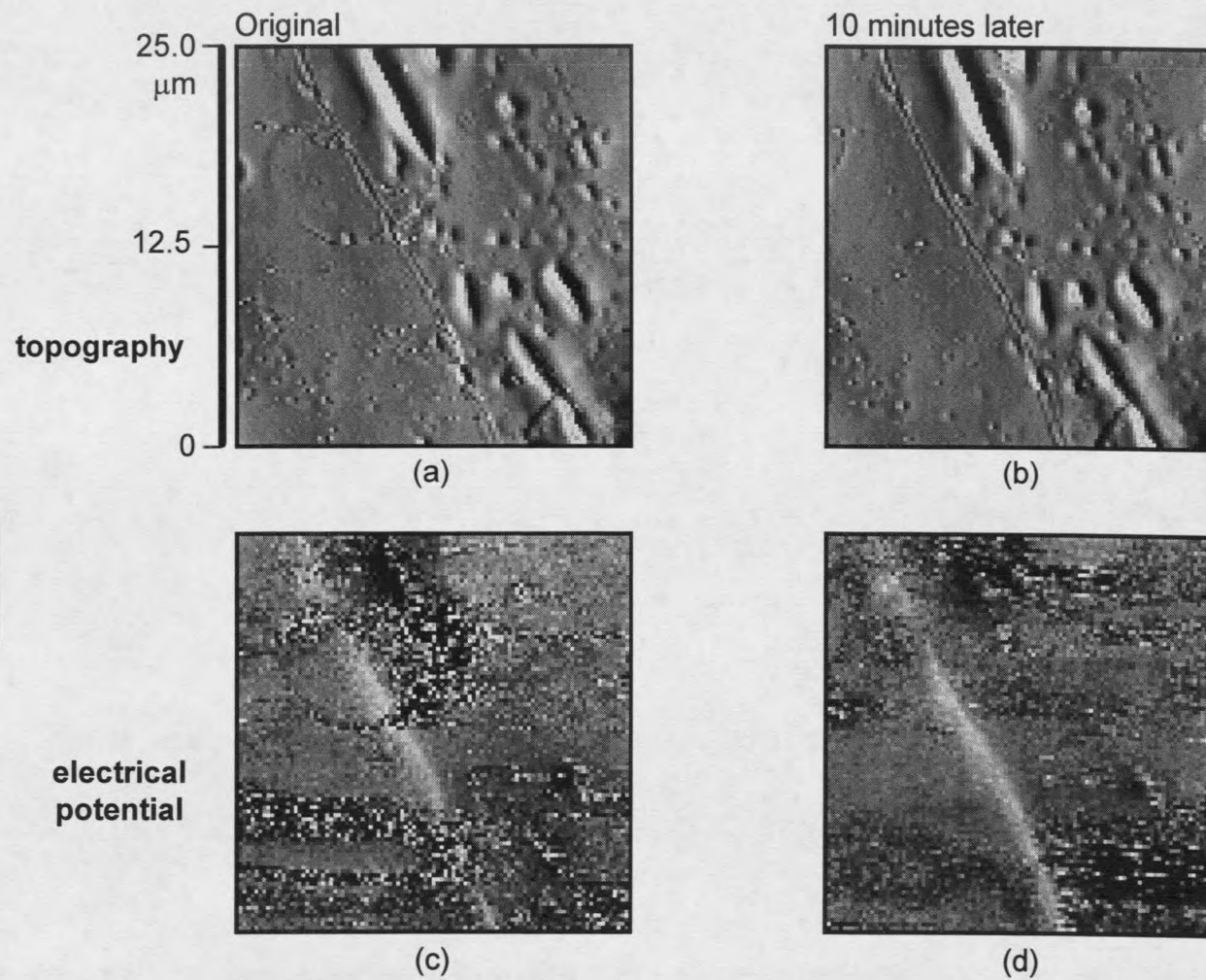


Figure 7.4. Time dependent studies of KFM on a PZT ceramic surface.

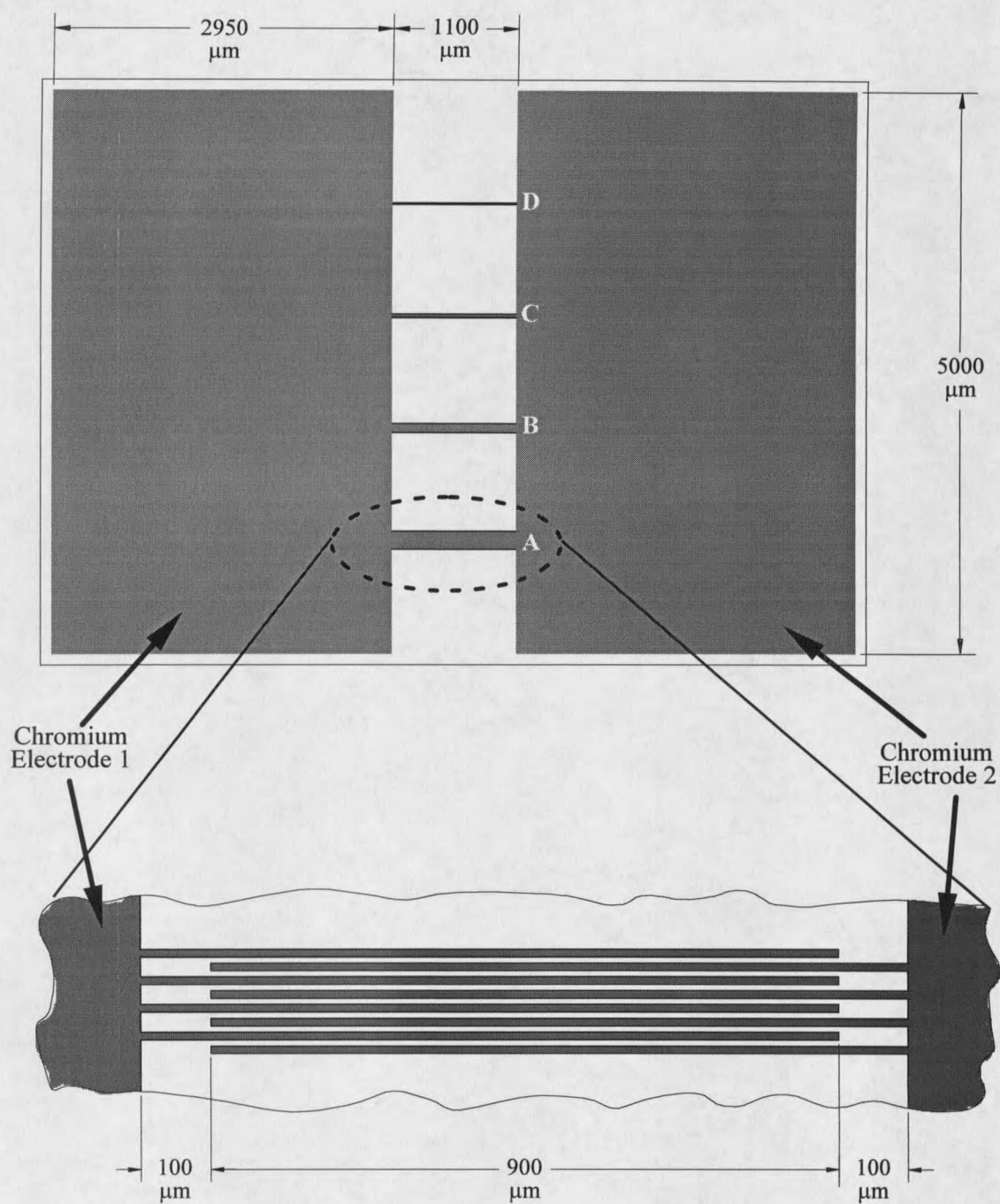


Figure 7.5. Design of the standard sample for electrical potential measurement.

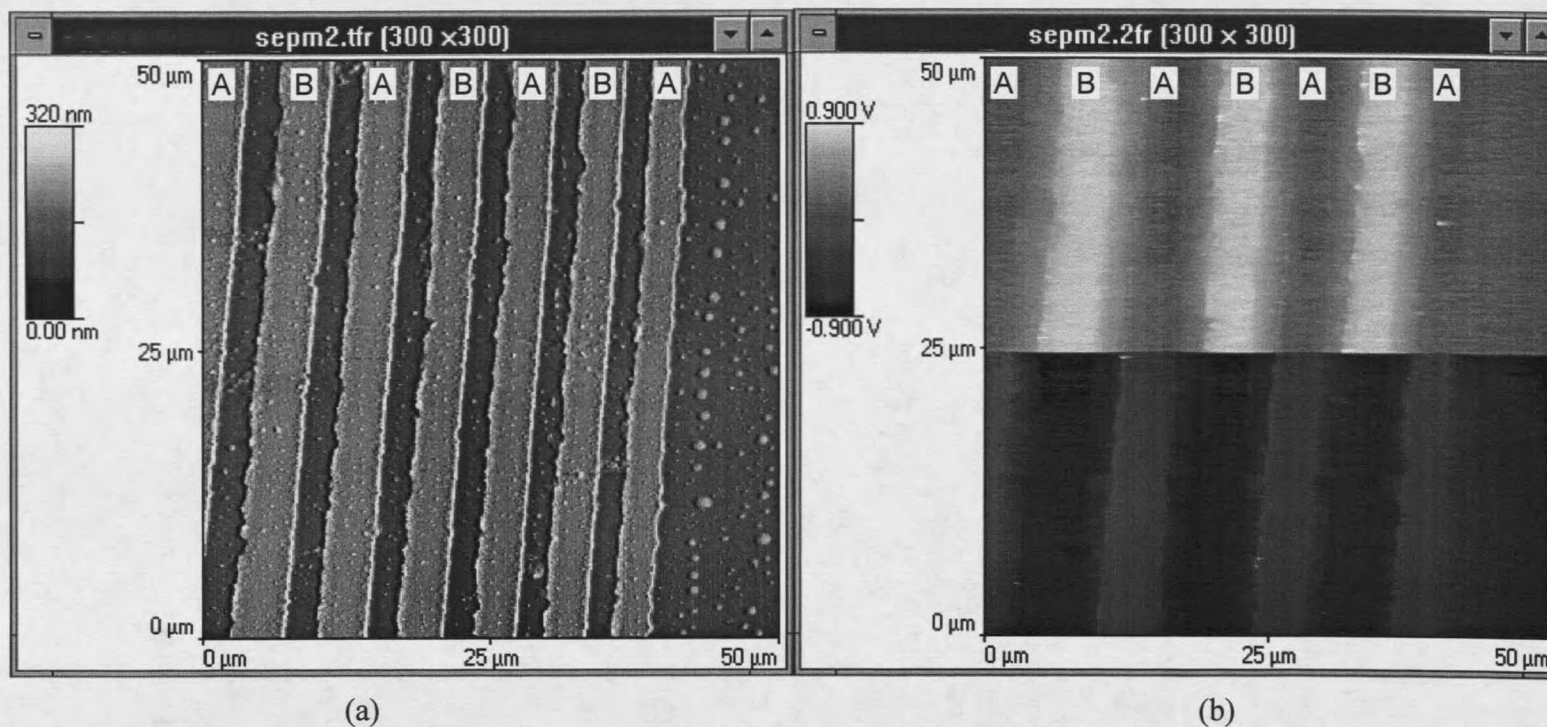


Figure 7.6. Simultaneous topographic (a) and surface potential (b) images of the standard sample. The electrodes labeled “A” were always connected to ground. The electrodes labeled “B” were connected to a voltage source controlled by the software. Each image has 300 scan lines. Each line has 300 data points.

The first problem to be studied is the possible crosstalk between the topographic feedback loop and the electrical potential feedback loop. If there is any crosstalk, it can be seen in the images. To help studying the crosstalk, the controlled voltage is changed from +1V to -1V in the middle of the scan. From the Figure 7.6(a), we can see that when the surface bias is switched from +1V to -1V the topographic image is not affected. This indicates that the change in surface potential does not affect the topographic feedback loop. On the other hand, let us consider the last electrode on the right of the image (labeled "A"). The topographic image shows a large step between the electrode and the substrate. However, since this electrode is grounded, the surface potential is similar to the substrate. In the Figure 7.6(b), the boundary between the last "A" electrode and substrate is not clear, which indicates that the change in surface topography has almost no effects on the electrical potential feedback loop. This can be further shown with a set of line profiles from the image set (Figure 7.7). Therefore, the two feedback loops can operate properly at the same time with no crosstalk.

The second problem to be studied is the accuracy of the potential measurement. In Chapter Three, it has been demonstrated that the temporal change of surface potential on a flat sample can be measured accurately. However, the measurement of spatial change in surface potential is a different issue. The standard sample was used to test the measurement of such spatial variation of the surface potential. From the images in Figure 7.6, we can analyze the result of the measurement. The electrodes labeled "A" were always grounded. The electrodes labeled "B" were connected to an external voltage source. Figure 7.8 shows three surface potential measurements on three different scan lines, where line No. 80 is in the middle of the region where electrodes "B" have +1 V bias, line No. 152 is the line right after the bias voltage on the electrodes "B" was switched to -1 Volt, and line No. 299 is the last line of the image.

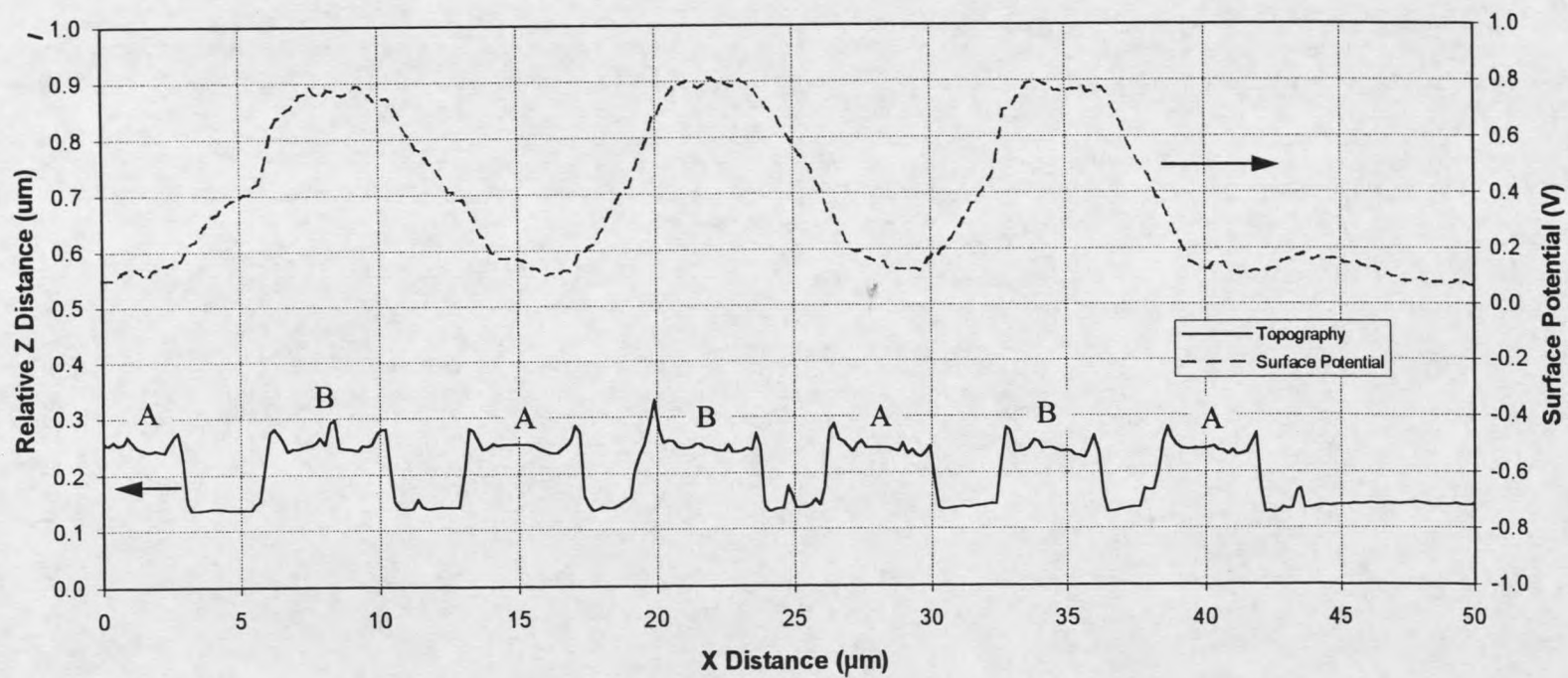


Figure 7.7. Line No. 80 of the images in Figure 7.6.

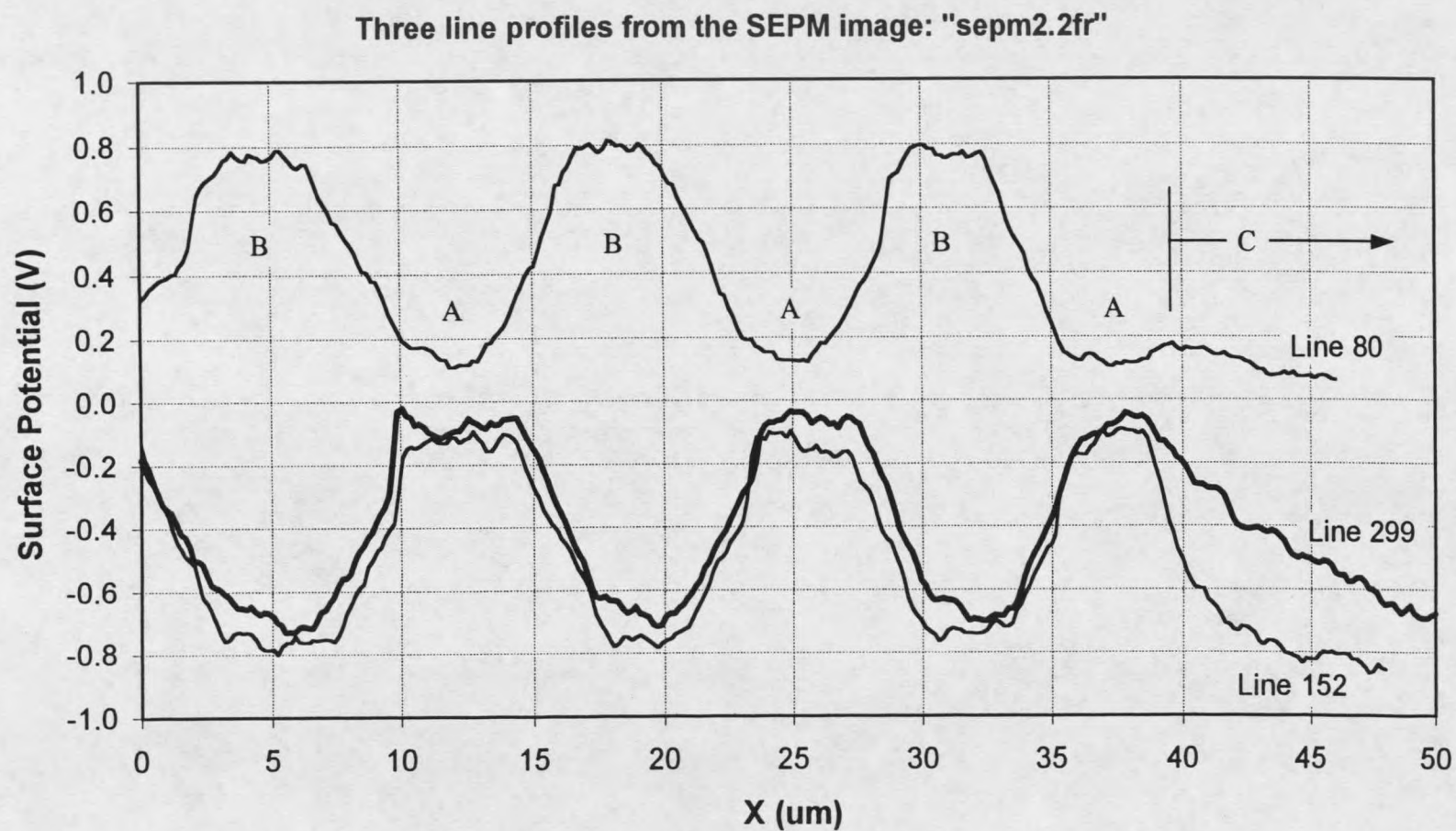


Figure 7.8. Three line profiles from the potential image show the relative of the potential changes.

Although the external voltage is only applied to one set of the electrodes, the spreading of the electric field reduces the localization of the surface potential. Since the tip is maintained at a specific distance from the surface, the measured potential difference between two sets of electrodes is about 0.7 volt. Comparing line No. 80 and No. 152 in Figure 7.8, when the voltage on electrodes "B" is switched from +1 V to -1 V, the potential on electrodes "A" is changed too. This indicates that the electric field from electrodes "B" extends to the space above electrodes "A". Therefore, the measurement of spatial changes in the surface potential depends on the average distance between the tip and the sample. The measurement results can also be affected by the amplitude of cantilever vibrations, and amplitude of the oscillating bias voltage.

From Figure 7.8, we can see the electrostatic effect of the surface contamination again. When localized charges exist on the surface, some opposite charges will get into the surface contamination layer to try to screen the unbalanced charge. This process can give some electrostatic characteristics to the contamination layer. Comparing line No. 80 and No. 152, the change on the right side of the last "A" electrode is the most significant. In region "C", the surface material is the intrinsic silicon. When electrodes "B" are at +1 volt, the surface of the sample is mostly negative charged. When the voltage on electrodes "B" is switched to -1 V, the negative charge in region "A" can be removed easily due to the grounding of electrodes "A". However, in region "C", since the intrinsic silicon has very small conductivity, the negative surface charge in the contamination layer can not be removed right away. This results in the appearance of a negative bias in region "C". This negative charge in region "C" is not stable, and will deplete with time. Comparing line No. 152 and No. 299, such change in electrostatics in the contamination layer is obvious. In line No. 299, region "C" has a potential closer to zero than in line No. 152.

In the above analysis, it is shown that the standard sample is important in studying the performance of the KFM. With the standard sample, the success of the operation of the double-

feedback-loop system is demonstrated with the measurement of both spatial and temporal changes of the surface potential. The measurement of spatial variation of surface potential is influenced by a number of parameters, such as tip-sample gap, cantilever vibration amplitude, oscillation amplitude of bias voltage, and so on. Further research is needed on this issue. Finally, this experiment further demonstrates the electrostatic properties of the surface contamination layer.

Studies of Sub-Micrometer MBE Grown Layers

To demonstrate the application of the KFM to solve problems, a cross-section sample of an MBE grown multilayer structure was imaged. On a semiconductor surface, the change of the band structure, due to the change in dopant density, can be measured as the changes of surface potential. For an MBE grown multilayer structure, the dopant concentration can be controlled to vary from layer to layer. When the sample is cleaved, the cross-section can be examined to measure the variation of dopant concentration in depth. Since the MBE layers are on the top of the wafer, the cross-section of the multilayer is on the edge of the sample. To locate an area of the multilayer, the combined AFM/SEM instrument is needed. A sample with half-micrometer MBE layers on a semiinsulating GaAs substrate was used to test this technique.

With STM, the measurements of dopant concentration have been done on heavily doped semiconductor samples. However, with intrinsic semiconducting/semiinsulating samples, or samples with thick surface oxide, the STM technique has a fundamental problem. Since STM has to use a tunneling current to detect the tip-sample gap, the sample has to have sufficient conductivity for establishing feedback. Because the substrate of our sample was an intrinsic semi-insulating GaAs, STM was tried but it failed. Since there was not enough tunneling current to maintain tunneling feedback, the STM tip crashed into the sample. Therefore, the STM technique is not appropriate for surface potential measurements on samples that do not have enough conductivity.

Since the KFM is mostly an AFM based on the VCM operation, it does not require any cur-

rent between the tip and the sample to maintain feedback. Therefore, surface potential changes can be measured on non-conductive samples. Since our sample was intrinsic semiinsulating GaAs, KFM was the only method available to measure the surface potential.

Sample Preparation

The MBE layers were grown on an intrinsic semiinsulating GaAs (100) substrate. The thickness of each layer was monitored by RHEED during the grow process, and all layers were controlled to be 500 nm thick.⁹² Figure 7.9 shows the layer structure of the sample. The sample was mounted on a special holder to allow imaging of the cross-section.

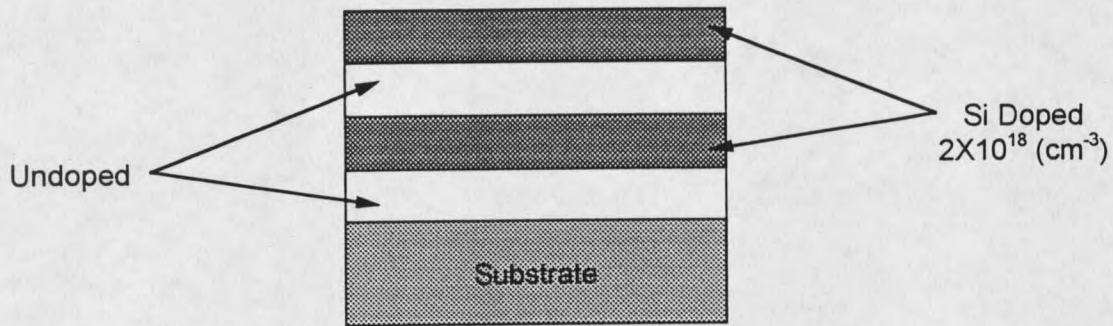
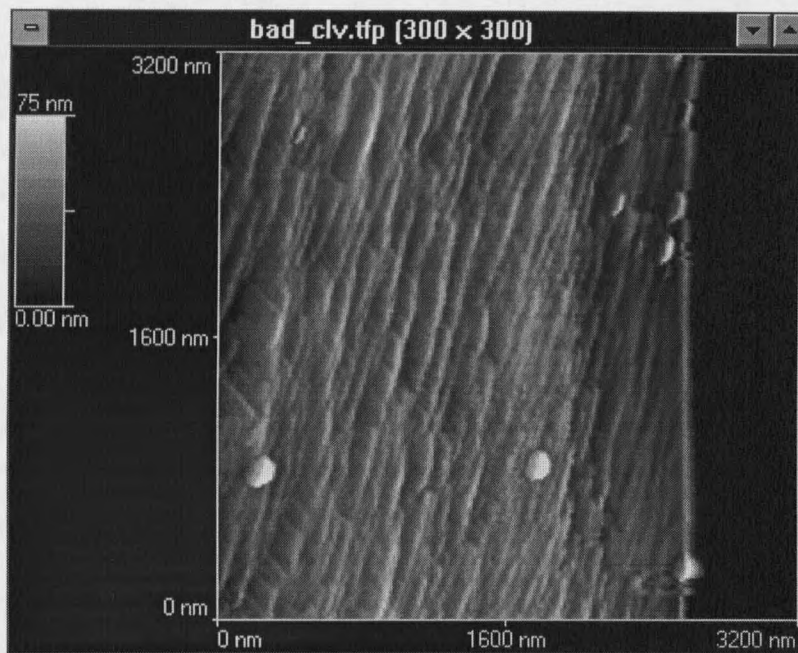
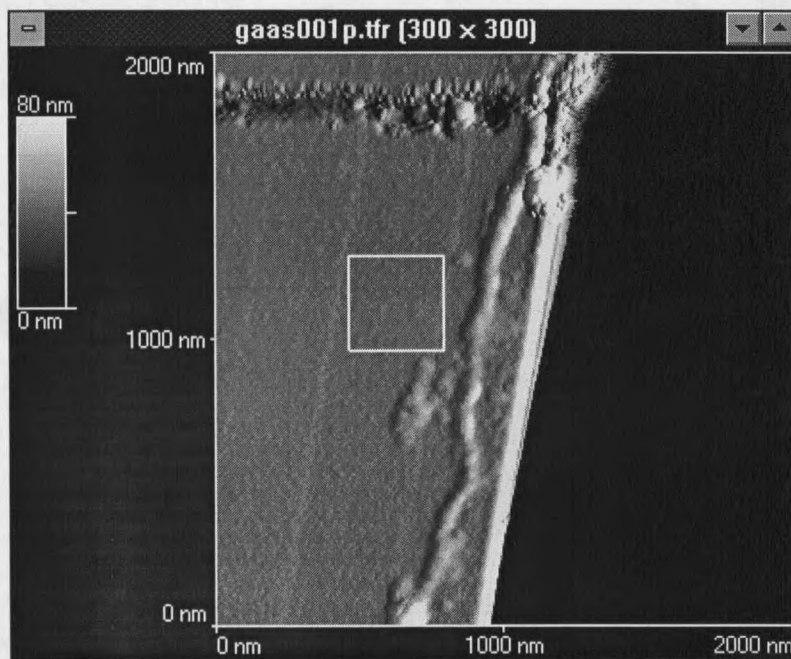


Figure 7.9. The GaAs MBE grown multilayer sample.

To examine the cross-section, the wafer needs to be cleaved first. The cleavage process can create defects on the surface of the cross-section that can influence the measurement of surface potential. To measure the bulk dopant concentration, the number of defects on the cross-section must be minimized. This requires a “clean” cleave. The quality of cleavage performed in air can be easily seen in the AFM topographic images shown in Figure 7.10. Originally, when the cleaving was done poorly, the result was a bad cleave which contained a large number of small atomic steps, as shown in Figure 7.10(a). Later, when the cleaving was improved, a large atomically flat surface can be obtained, which is clearly shown in Figure 7.10(b).



(a)



(b)

Figure 7.10. The surface topography of cleavage surfaces. (a) a bad cleave. (b) a good cleave.

After cleaving the sample properly, some samples were left in air to be naturally passivated, while others were immediately followed by $(\text{NH}_4)_2\text{S}$ passivation;⁹³ the samples were kept in the $(\text{NH}_4)_2\text{S}$ solution for 10 minutes. The cleavage surface looked very good after $(\text{NH}_4)_2\text{S}$ passivation. The AFM topographic image shows large sections of atomically flat surfaces (Figure 7.10(b)). The surface roughness on a single atomic step, indicated by the square box in Figure 7.10(b), is 0.34 nm (area RMS roughness).

Locating the Area of MBE Layer

Since the cross-section of the multilayer is within a 2 μm region from the edge of the sample, the combined AFM/SEM instrument was used to position the tip to the multilayer area. The sample was mounted on a sample clamp, with the cross-section surface facing up, and then was introduced into the SEM through the load-lock chamber. The SEM was used to guide the tip to a position over the area of multilayers. Originally, the tip was placed over the substrate. Then, while monitoring with the SEM, the primary translator was controlled to move the multilayer region under the tip. To minimize the effect of electron beam damage, low electron beam energy, 2 keV, was used. Figure 7.11 shows the final positions of the tip and the sample.

Surface Potential Imaging

Because of the difficulty of controlling two feedback loops in vacuum, surface potential imaging was performed in air. After positioning the tip to the area of MBE layers under the SEM, the AFM system was carefully moved from the SEM specimen chamber into the load-lock chamber. Then, the load-lock chamber was vented to air, and a pair of images of topography and surface potential was simultaneously acquired (Figure 7.12). The surface of the cross-section of the MBE layers was passivated in air. In the surface potential image, three boundaries can be clearly seen. They are the boundaries that separate the four MBE grown layers. A pair of line profiles of the cross-section is also shown in Figure 7.12.

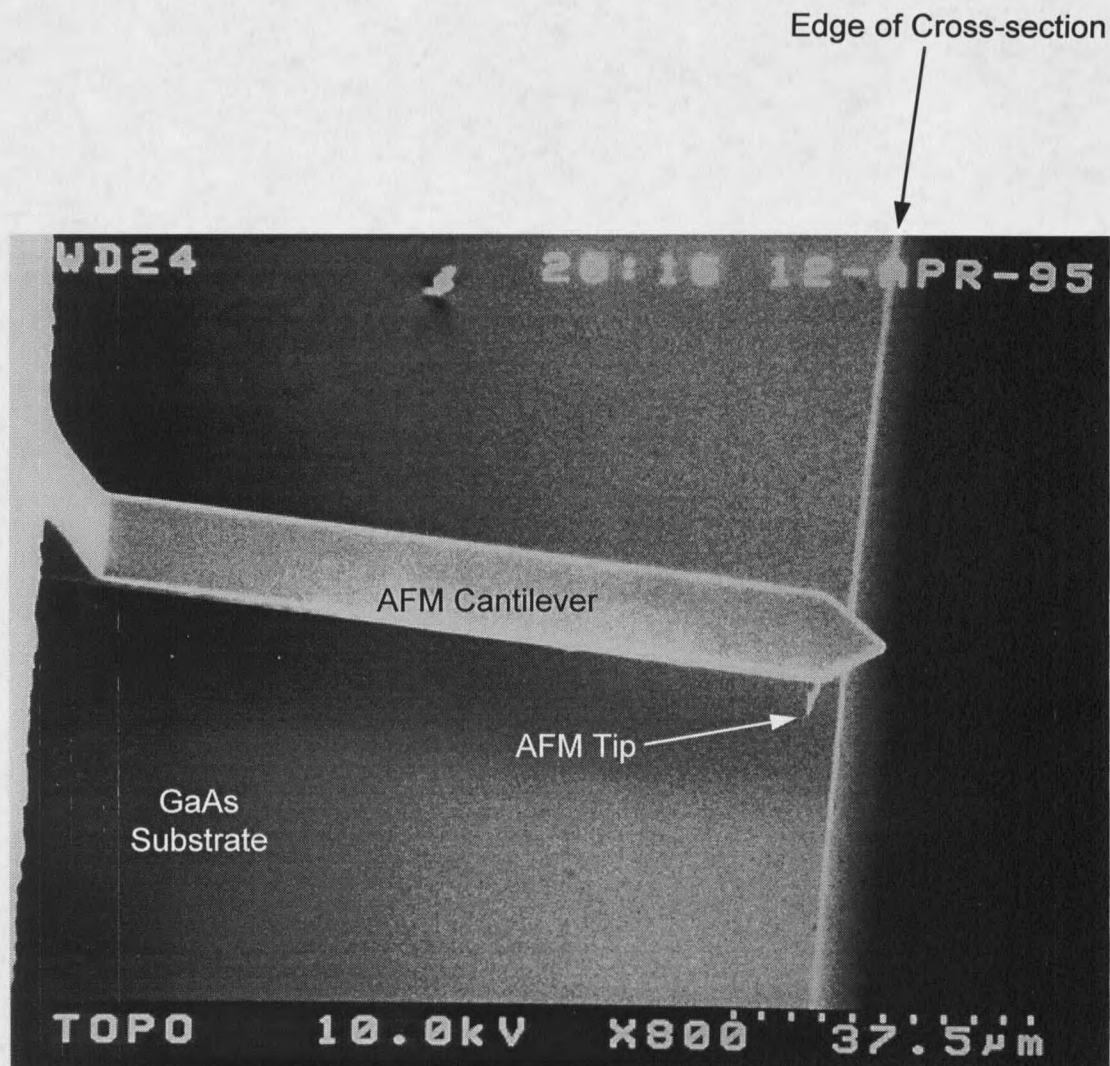


Figure 7.11. SEM view showing positioning of the AFM tip over the area of MBE layers.

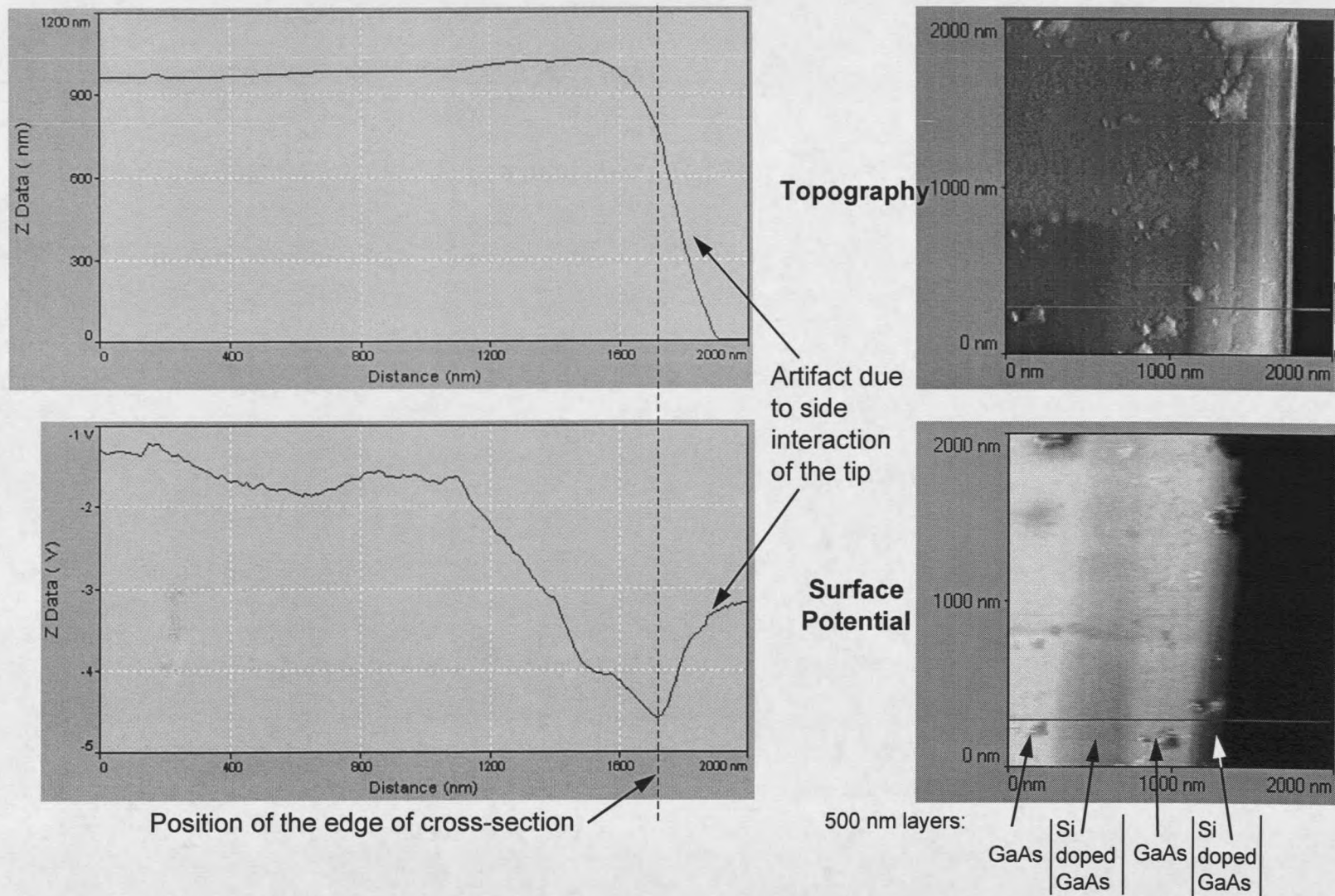


Figure 7.12. Simultaneous topographic and surface potential images of the MBE layers.

Results and Analysis

The purpose of a cross-section study is to obtain information in the bulk. If the surface of a cross-section is uniform and free of defects, the surface potential measured on a cross-section can correspond to the energy band structure in the bulk. However, at the edge of the cross-section, which corresponds to the surface of the wafer, the surface potential will no longer resemble the bulk.

The first outstanding feature in the Figure 7.12 is the edge of the cross-section. From the line profile of topography, it is hard to locate where the edge is. The geometric convolution between the tip (with finite aspect ratio) and the sample makes the sharp edge disappear from the image. However, in the line profile of the surface potential, a potential minimum is clearly seen, which is the position of the edge. For the potential image, the electrostatic interaction is mostly at the tip, which makes a significant difference between the case of tip-on-top and the case of tip-drop-off. As the tip drops off more, it gets further from the sample and the interaction gets weaker, which results in the measured surface potential approaching zero.

The edge itself is a huge defect for the cross-section surface. Also, from the topographic image, we can see the surface near the edge has a lot of steps. At the edge, many new surface states exist for the carrier. Since the silicon-doped layer is n-type, the surface states are mostly filled up by electrons. From the line profile, we can see the last layer has comparably stronger negative potential, which even influences the subsequent layers.

Since the edge is so hard to predict, we shall ignore the measurement on the last MBE layer that is on the top of the wafer. The KFM image of the cross-section of MBE layers in Figure 7.12 shows the changes in the surface potential across the layers. A line profile covering three MBE layers is taken from the surface potential image (see Figure 7.13), which covers two interfaces, from the undoped region (A) to the doped region (B), and from (B) to the undoped region (C).

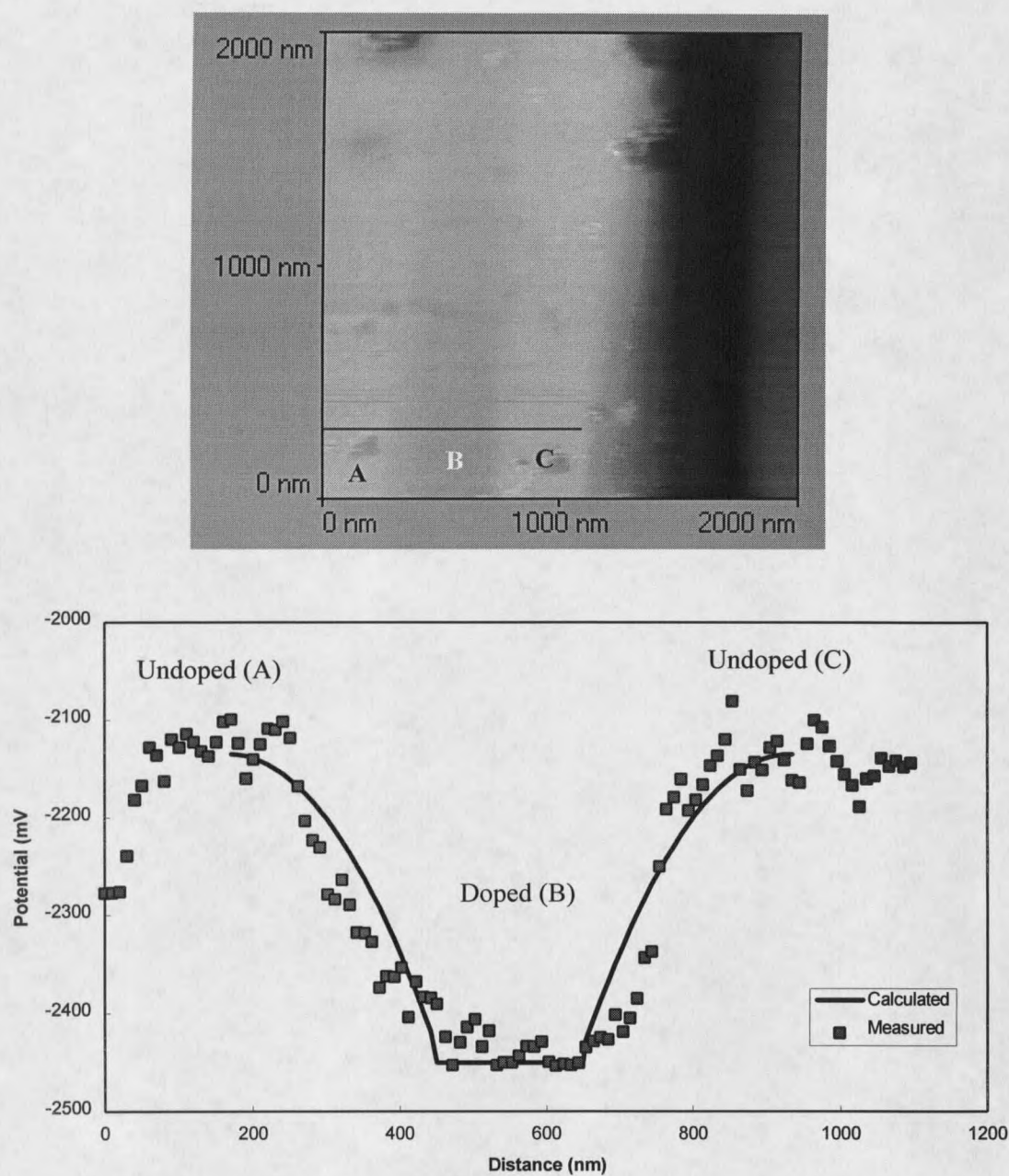


Figure 7.13. A line profile taken from the surface potential image. (a) The location of the line in the KFM image; (b) Comparing the measured data with the calculated data.

Since there is an initial difference of Fermi level ($\Delta\Phi$) between the doped and undoped region, charges will transfer across the interface to achieve a steady state. The free carrier will be depleted from the volume near the interface. Assuming the charge distribution is uniform in the depletion layer, we have the following surface potential change across the interface. From A to B,

$$\phi_{AB}(x) = \begin{cases} \Delta\Phi - \frac{eN_{ND}}{2\epsilon\epsilon_0}(x - x_{AB} + w_{ND})^2 & x_{AB} - w_{ND} < x < x_{AB} \\ \frac{eN_D}{2\epsilon\epsilon_0}(x - x_{AB} - w_D)^2 & x_{AB} < x < x_{AB} + w_D \end{cases} \quad (7.1)$$

From B to C,

$$\phi_{BC}(x) = \begin{cases} \frac{eN_D}{2\epsilon\epsilon_0}(x - x_{BC} + w_N)^2 & x_{BC} - w_D < x < x_{BC} \\ \Delta\Phi - \frac{eN_{ND}}{2\epsilon\epsilon_0}(x - x_{BC} - w_{ND})^2 & x_{BC} < x < x_{BC} + w_{ND} \end{cases} \quad (7.2)$$

where N_{ND} is the impurity concentration in the undoped region, and N_D is the dopant concentration of the silicon doped region. The fitting curve shown in Figure 7.13 was calculated by setting $N_{ND} = 5 \times 10^{15} \text{ cm}^{-3}$, $N_D = 2 \times 10^{18} \text{ cm}^{-3}$, $\Delta\Phi = 300 \text{ mV}$, $x_{AB} = 450 \text{ nm}$, and $x_{BC} = 650 \text{ nm}$.

In general, the above fitting is good. The obvious problem with this fitting is the unnaturally sharp corner on both sides of the doped region. The reason for this is the assumption of linear charge distribution, which is only a crude approximation. In reality the charge distribution is changed gradually. If such a model includes the gradual change of the charge distribution, the theoretical surface potential should change smoothly across the interface.

Conclusion

The scanning Kelvin-probe force microscope (KFM) is a surface potential imaging tool based on the classical Kelvin probe technique, which can image spatial variation of surface electrical potential without any conductivity requirement on the samples. It contains two feedback loops that

operate simultaneously to measure surface topography and electrical potential.

We designed and built the first standard sample for examining the performance of this surface potential imaging tool. With this sample, we showed that the two feedback loops can be operated with almost no crosstalk.

We enhanced the original method by studying the temporal information of KFM images. On a piezoelectric sample, we observed fluctuation of surface potential that varies dramatically with time. This phenomenon could be caused by a patch of space charge existing within the surface contamination. Because of the liquid-like property of the contamination layer, the space charges can float around due to the changes of environment.

We demonstrated an application of KFM in measuring the dopant concentration on a semi-insulating GaAs cross-sectional sample that is not applicable to STM because of the lack of sample conductivity. The surface potential changes due to the change of the dopant concentration in the MBE-grown multilayer structure were measured. The calculated dopant concentrations from the measured potential are close to the values obtained from the MBE growth process.

With the combined PPM/SEM instrument, we were able to measure the surface potential at the edge of a surface for the first time, where we observed the strong edge effect on the surface potential image near the edge of the cross-section. This is due to the intrinsic long-range characteristics of electrostatic interaction. A theoretical model of electrostatic interaction between a cone-shape conductor and the surface edge is needed to describe this edge effect which could lead to a measurement of effective tip size.

CHAPTER 8

CONCLUSIONS

The research of this thesis has been focused on the studies of nanometer scale tip-sample interactions by using the nanometer scale capabilities of the proximal probe microscope (PPM), especially the cantilever-based atomic force microscope (AFM). To explore new experimental territories, new experimental apparatus and methods were developed. With these new experimental capabilities, new insights were obtained on surface contamination layers, nanometer scale capillary force, surface electrostatic properties, nanometer scale surface contact, and the working mechanism of vibrating cantilever AFM.

Proximal Probe Microscopy

The method of using a material probe positioned in close proximity to a surface to obtain microscopic images is called proximal probe microscopy (PPM). The scanning proximal probe microscope that is based on microscopic force sensors is called the scanning force microscope (SFM). Compared to the scanning tunneling microscope (STM), SFM has been defined as a technique that "no longer uses electron tunneling to probe the local properties of sample surfaces, but rather the tip-sample force interaction".⁹⁴ The earliest force sensor was based on a microscopic cantilever, using its spring characteristics to measure the force. This cantilever-based force microscope has been named the atomic force microscope (AFM). "The spring in the AFM is a critical component."⁹¹ The research of this thesis is in the field of SFM. The following diagram illustrates the relationship of all the SFM modes.

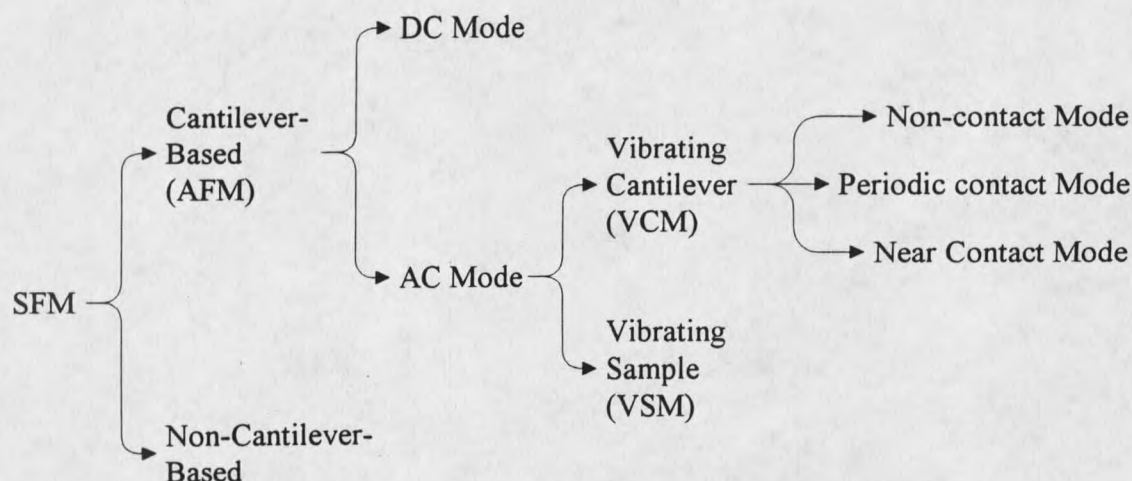


Figure 8.1. Classification of various SFM modes.

Advances in the field of microscopy have been marked by the continuous increase of spatial resolution. In the era of optical microscopy (far field), Abbe⁹⁵ pointed out that the ultimate resolution is limited by the wavelength of radiation used. However, this concept was challenged later by Synge,⁹⁶ who noted that resolution was not limited by wavelength when a tiny light source smaller than the wavelength is used and the distance between the light source and sample is smaller than the wavelength. This was the first idea for the novel near-field microscope, which indicated that the breakthrough of spatial resolution must come from near-field imaging. The PPM is the most recent advance of microscopy due to its capability of atomic resolution. When making the analogy between the tip of a PPM probe and an optical lens and considering the tip-sample gap in the nanometer scale, then the PPM is considered a near-field microscope. The near-contact mode described in this dissertation for achieving maximal capability of the image resolution of AFM is developed under the same guidelines as near-field imaging.

Experimental Apparatus and Methods

Generally, as the spatial resolution of a microscope increases, the image field-of-view de-

creases. For a microscope to be practical, there must be a method to move the image field-of-view to the area of interest on the sample surface. To locate sub-micrometer surface features, a PPM was built and integrated with a SEM, using the SEM to locate the area of interest. This PPM includes both an STM and an AFM with a light-lever deflection detection force sensor. The PPM can be operated simultaneously with the SEM. Two levels of XYZ translation stages were built, to move the PPM tip to the center of SEM field-of-view and precisely position the sample under the PPM tip. This allows easy location of sub-micrometer scale features on a large sample.⁹⁷ The O-ring-mass vibration isolation setup, along with the vacuum chamber, provides a very quiet environment for PPM operation.

In addition to its use as a ranging tool, the SEM is used to monitor the condition of the PPM tips, which helps to ensure the reliability of PPM images. With this instrument, the conditions of PPM tips were studied for developing the optimal operation condition in the vibrating cantilever mode (VCM). SEM analysis showed that the tips were obviously damaged after operating in periodic contact condition of the VCM.

A method of studying the properties of the surface contamination layer was developed, using the capability of the new AFM's working environment, which can be changed between ambient air and vacuum. Under ambient air conditions, a layer of residual contamination always remains on the surface of a sample. Under vacuum, the liquid-like contamination layer is removed.

In the loading curve measurement, an analysis method was developed to remove cantilever deflection in the raw data. With this correction, the loading curves can be plotted against the tip-sample gap, which make it readily comparable to any theoretical model.

Surface Contamination and Capillary Force

The surface contamination layer is thought to apply a capillary pulling force on AFM tips. The magnitude of this attractive force was thought to be stronger than all types of cantilevers cur-

rently available, creating the jump-to-contact (JTC) condition. When the JTC occurs, the AFM tip will be captured by the contamination layer, and sustains damage. Loading curve measurements show that when the spring constant of a cantilever is great enough, the JTC does not occur. On a particular aluminum mirror sample, the threshold spring constant was about 50 N/m, which indicates that it is possible to prevent contact damage of the tips in the VCM by using cantilevers of sufficiently larger spring constant.

The surface contamination layer contributes significantly to the fluctuation of the loading curves measurement. The measurement results are more consistent in vacuum because the contamination is mostly removed. A set of loading curves was measured with the same tip on the same sample area in both air and vacuum. Fitting the approach curve in air with a theoretical model of capillary force, the thickness of the contamination layer was estimated to be about 6 nm. In this case, the magnitude of the capillary force was 230 nN.

Optimal Operation Conditions for the Vibrating Cantilever Mode AFM

When operating VCM in air, three different working conditions can be distinguished. The classification can be made based on the relationship between the tip, the surface contamination layer, and the sample:

- 1) Traditional non-contact condition: In this mode, the tip stays near the top of the contamination layer.
- 2) Near-contact condition: In this mode, the tip vibrates within the contamination layer. It moves into the bottom of the contamination layer, and stays close to the sample surface without direct physical contact.
- 3) Periodic contact condition: In this mode, the cantilever vibration amplitude is relatively large (usually ≥ 100 nm). The tip moves in and out of the contamination layer. The tip makes direct physical contact to the sample at the bottom of every oscillation.

Using the surface contamination layer has a substantial advantage in preventing tip and sample damage. The molecular layer that bonds directly to the sample and the tip surfaces is very hard to remove. Because of the existence of this last chemisorpt layer, a tip with a small vibration amplitude can be brought very close to the surface without significant damage. When the vibration amplitude is small, the energy stored in the vibrating cantilever is equivalent to the chemisorpt energy of the surface molecular layer. Therefore, the maximum possible damage in the near-contact mode can only happen to this surface molecular layer.

In the periodic contact condition, tip damage was observed. Two mechanisms for probe geometry changes were discovered. In the first mechanism, the probe changes geometry by picking up contamination from the sample surface. In the second mechanism, the probe is physically worn off by contacting the surface. Both of these mechanisms cause substantial reduction in lateral resolution.

The image resolution increases as the tip-sample gap decreases. When the tip is in the near-contact condition, high-resolution images can be acquired without tip and sample damage. Therefore, the near-contact mode is the optimal working condition for VCM operation. This mode can be achieved by using a cantilever with a large spring constant, driving the cantilever with a small vibration amplitude, and choosing a low set point.

Nanometer Scale Surface Potential Measurement

While the AFM VCM operation maintains a constant average tip-sample gap, the electrical potential of the surface can be measured by the Kelvin method, which is the concept of scanning Kelvin-probe force microscopy (KFM). This surface potential imaging capability, which is a variation of VCM operation, was implemented on the new combined PPM/SEM instrument. It is a double feedback loop system, where the first loop is the ordinary VCM for tracking surface topography, and the second loop is used to measure surface potential with the Kelvin method. Although

this technique does not provide atomic resolution like the STM, it can image samples that are not conductive enough for the STM. A standard sample was developed to test the performance of this surface potential imaging system.

With KFM, the fast time dependence of surface potential was observed on a PZT piezoelectric ceramic sample, which confirmed that the surface contamination contains free charges to balance the intrinsic charge on the highly polarized piezoelectric surface.

The surface of a cross-section of GaAs MBE grown layers on a seminsulating substrate was studied. This is a sample not workable with STM due to the lack of conductivity. The surface potential variation caused by the variation of the doping concentration was measured. The measured surface potential was compared with the theoretical model, and the impurity concentration was determined from the comparison.

Future Studies

For the combined AFM/SEM instrument, it is important to understand the possible interference between the two microscopes. Studies should be made to understand the mechanism of the additional force applied to the AFM cantilever by the electron beam. This additional force could be caused by the momentum transfer from the electrons and/or the localized static charging of the cantilever. The AFM may be able to analyse the electron-beam induced surface contamination.

Future experiments on surface contamination layers should be performed under controlled humidity.⁵ The relationship between the layer thickness and nanometer scale capillary force should be measured. For small amplitude VCM operation, the transition from near-contact to physical contact should be investigated. The existence of a tightly bonded molecular layer on the bottom of the contamination can be confirmed by comparing the tip-sample interactions on surfaces of polar material with non-polar material. A quantitative theoretical model, involving the specific structure of the surface contamination layer on both the tip and the sample, needs to be developed for understanding the nanometer-scale tip-sample interaction in air.

REFERENCES CITED

- ¹ G. Binnig, C. F. Quate, and Ch. Gerber, Phys. Rev. Lett. **56**, 930 (1986).
- ² T. R. Albrecht and C. F. Quate, J. Vac. Sci. Technol. A **6**, 271 (1988).
- ³ T. R. Albrecht and C. F. Quate, J. Appl. Phys. **62**, 2599 (1987).
- ⁴ H. K. Wickramasinghe, J. Vac. Sci. Technol. A **8**, 363 (1990).
- ⁵ G. Yang, J. P. Vesenka, and C. J. Bustamante, Scanning, **18**, 344 (1996).
- ⁶ Y. Martin, C. C. Williams, and H. K. Wickramasinghe, J. Appl. Phys. **61**, 4723 (1987).
- ⁷ U. Landman, W. D. Luedtke, N. A. Burnham, and R. J. Colton, Science, **248**, 454 (1990).
- ⁸ Q. Zhong, D. Innis, K. Kjoller, and V. B. Elings, Surface Sci. Lett. **290**, L688 (1993).
- ⁹ S. C. Meepagala, F. Real, and C. B. Reyes, J. Vac. Sci. Technol. B **9**, 1340 (1991).
- ¹⁰ N. A. Burnham, R. J. Colton, and H. M. Pollock, J. Vac. Sci. Technol. A **9**, 2548 (1991).
- ¹¹ H. J. Ho and P. E. West, U. S. Patent #5,455,420 (October 1995).
- ¹² H. Ho and P. West, Scanning, **18**, 339 (1996).
- ¹³ E. J. Abbott, S. Bousky, and D. E. Williamson, Mechanical Eng., 205 (March 1938).
- ¹⁴ R. Young, J. Ward, and F. Scire, Rev. Sci. Instrum. **43**, 999 (1972).
- ¹⁵ G. Binnig, H. Rohrer, Ch. Gerber, and E. Weibel, Phys. Rev. Lett. **49**, 57 (1982).
- ¹⁶ H. K. Wickramasinghe and D. Rugar, IBM J. Res. Develop. **39**, 602 (1995).
- ¹⁷ The traditional optical microscopes are *far-field* microscopes, where the distance between the objective lens and sample is greater than the wavelength of their probes (photons). In this text, the *near-field* condition means that the distance between the tip of a PPM probe and sample is smaller than the wavelength of light.

- ¹⁸ Due the nonlinearity characteristics of the piezoelectric ceramics, various types of independent position encoders were developed to measure the surface topography in higher degree of accuracy.
- ¹⁹ D. P. E. Smith, *Rev. Sci. Instrum.*, **66**, 3191 (1995).
- ²⁰ T. Goddenhenrich, H. Lemke, U. Hartmann, and C. Heiden, *J. Vac. Sci. Technol. A* **8**, 383 (1990).
- ²¹ C. M. Mate, G. M. McClenlland, R. Erlandsson, and S. Chiang, *Phys. Rev. Lett.* **59**, 1942 (1987).
- ²² G. Meyer and N. M. Amer, *Appl. Phys. Lett.* **53**, 1045 (1988).
- ²³ M. Tortonese, R. C. Barrett, and C. F. Quate, *Appl. Phys. Lett.* **62**, 834 (1993).
- ²⁴ J. W. P. Hsu, M. Lee, and B. S. Deaver, *Rev. Sci. Instrum.* **66**, 3177(1995).
- ²⁵ K. Karrai and R. D. Grober, *Appl. Phys. Lett.* **66**, 1842(1995).
- ²⁶ For example, a STM system manufactured by QuanScan, Pasadena, CA (1989).
- ²⁷ Ch: Gerber, G. Binnig, H. Fuchs, O. Martin, and H. Rohrer, *Rev. Sci. Instrum.* **57**, 221 (1986).
- ²⁸ P. M. Thibado, Y. Liang, and D. Bonnell, *Rev. Sci. Instrum.* **65**, 3199 (1994).
- ²⁹ D. A. Walters, D. Hampton, B. Drake, H. G. Hansma, and P. K. Hansma, *Appl. Phys. Lett.* **65**, 787 (1994).
- ³⁰ A. V. Ermakov and E. L. Garfunkel, *Rev. Sci. Instrum.* **65**, 2853 (1994).
- ³¹ U. Stahl, C. W. Yuan, A. L. de Lozanne, and M. tortonese, *Appl. Phys. Lett.* **65**, 2878 (1994).
- ³² G. J. Collins and H. Ho, *Microscopy Today*, No. 95-2, p. 22 (1995).
- ³³ H. Becker, et. al., U. S. Patent #2,728,222 (1955) and D. Lee, U. K. Patent #2009409 (1978).
- ³⁴ T. R. Albrecht, P. Grutter, D. Horne, and D. Dugar, *J. Appl. Phys.* **69**, 668 (1991).
- ³⁵ F. J. Giessibl, *Science*, **267**, 68 (1995).
- ³⁶ S. Kitamura and M. Iwatsuki, *Jpn. J. Appl. Phys.* **34**, L145 (1995).
- ³⁷ H.Ueyama, M. Ohta, Y. Sugawara, and S. Morita, *Jpn. J. Appl. Phys.* **34**, L1086 (1995).
- ³⁸ N. W. Ashcroft and N. D. Mermin, *Solid State Physics*, Holt, Rinehart and Winston, pp.361 (1976).

- ³⁹ M. Nonnenmacher, M.P. O'Boyle, and H.K. Wickramasinghe, *Appl. Phys. Lett.* **58**, 2921 (1991).
- ⁴⁰ J. M. R. Weever and D. W. Abraham, *J. Vac. Sci. Technol. B* **9**, 1559 (1991).
- ⁴¹ H. Yokoyama, K. Saito, and T. Inoue, *Mol. Electron. Bioelectron.* **3**, 79 (1992).
- ⁴² J. N. Israelachvili, *Intermolecular and Surface Forces*, Academic Press, p. 8 (1992).
- ⁴³ *Ibid.*, p. 10.
- ⁴⁴ N. W. Ashcroft and N. D. Mermin, *Solid State Physics*, Holt, Rinehart and Winston, p. 398 (1976).
- ⁴⁵ J. N. Israelachvili, *Intermolecular and Surface Forces*, Academic Press, p. 113 (1992).
- ⁴⁶ *Ibid.*, p. 177.
- ⁴⁷ *Ibid.*, p. 176.
- ⁴⁸ *Ibid.*, p. 332.
- ⁴⁹ *Ibid.*, p. 178.
- ⁵⁰ *Ibid.*, p. 315.
- ⁵¹ C. J. Chen, *Introduction to Scanning Tunneling Microscopy*, Oxford University Press, p. 9 (1993).
- ⁵² J. D. Jackson, *Classical Electrodynamics*, John Wiley & Sons, Inc., p. 57 (1975).
- ⁵³ C. Kittle et al., *Mechanics* (Berkeley physics course, v. 1), McGraw-Hill, Inc. p. 216 (1973).
- ⁵⁴ *Ibid.*, p. 221.
- ⁵⁵ J. R. Hook and H. E. Hall, *Solid State Physics*, John Wiley and Sons, p. 173 (1985).
- ⁵⁶ Semiconductor Industry Association, *The National Technology Roadmap for Semiconductors*, p. 11 (1994).
- ⁵⁷ R. H. Brigham and S. M. Baumann, *Proc. Seventh Internat. Sym. Silicon Mat. Sci. Technol.* **94-10**, 277 (1994).
- ⁵⁸ G. Meyer and N. M. Amer, *Appl. Phys. Lett.* **53**, 1045 (1988).
- ⁵⁹ C. J. Chen, *Introduction to Scanning Tunneling Microscopy*, Oxford University Press, p. 224 (1993).

- ⁶⁰ G. Meyer and N. M. Amer, *Appl. Phys. Lett.* **56**, 2100 (1990).
- ⁶¹ R. C. Gamble, P. E. West, U. S. Patent #5,319,960 (1994).
- ⁶² J. H. Moore, C. C. Davis, and M. A. Coplan, *Building Scientific Apparatus*, Addison-Wesley Publishing Company, p. 41 (1983).
- ⁶³ Micro Pulse Systems Inc., Santa Barbara, CA 93110.
- ⁶⁴ Melles Griot 1995/96, p. 48-6 (1995).
- ⁶⁵ M. T. Postek, *Scanning*, **18**, 269 (1996).
- ⁶⁶ Hitach, *Instruction Manual for Model S-4100 Field Emission Scanning Electron Microscope*, Hitachi, Ltd. p. 3-42 (1991).
- ⁶⁷ M. E. Lines and A. M. Glass, *Principles and Applications of Ferroelectrics and Related Materials*, Clarendon Press, p. 12 (1977).
- ⁶⁸ Li Jingde, Lei Deming, and He Di, *Ferroelectrics*, **70**, 7(1986).
- ⁶⁹ C. J. Chen, *Introduction to Scanning Tunneling Microscopy*, Oxford University Press, p. 227(1993).
- ⁷⁰ P. Horowitz and W. Hill, *The Art of Electronics*, Cambridge University Press, p. 641 (1989).
- ⁷¹ *Ibid.*, p. 7.
- ⁷² *Artifacts in SPM*, TopoMetrix Corp., p. 7 (1993).
- ⁷³ E. I. Givargizov, *J. Vac. Sci. Technol. B* **11**, 449 (1993).
- ⁷⁴ E. I. Givargizov, A. N. Kiselev, L. N. Obolenskaya, and A. N. Stepanova, *Appl. Surf. Sci.* **67**, 73 (1993).
- ⁷⁵ J. H. Moore, C. C. Davis, and M. A. Coplan, *Building Scientific Apparatus*, Addison-Wesley, p. 36 (1983).
- ⁷⁶ D. Sarid, *Scanning Force Microscopy*, Oxford University Press, p. 13 (1991).
- ⁷⁷ A good method to remove the Z piezo artifact in loading curve measurement is to use an independent position encoder to measure the Z piezo movement.
- ⁷⁸ M. Binggeli and C. M. Mate, *Appl. Phys. Lett.* **65**, 415 (1994).

- ⁷⁹ H. A. Mizes, K.-G. Loh, R. J. D. Miller, S. K. Ahuja, and E. F. Grabowski, *Appl. Phys. Lett.* **59**, 2901 (1991).
- ⁸⁰ H. A. Mizes, private communication (1995).
- ⁸¹ S. P. Jarvis, et al., *Rev. Sci. Instrum.* **64**, 3515 (1993).
- ⁸² C. M. Mate, M. R. Lorenz, and V. J. Novotny, *IEEE Transactions and Magnetism*, **26**, 1225 (1990).
- ⁸³ J. Hu, X. D. Xiao, D. F. Ogletree, and M. Salmeron, *Science*, **268**, 267 (1995).
- ⁸⁴ H. K. Wickramasinghe, *J. Vac. Sci. Technol. A* **8**, 363 (1990).
- ⁸⁵ M. T. Postek and A. E. Vladar, *SPIE* **2725**, 504 (1996).
- ⁸⁶ K. L. Westra, A. W. Mitchell, and D. J. Thomson, *J. Appl. Phys.* **74**, 3608 (1993).
- ⁸⁷ U. Landman, W. D. Luedtke, N. A. Burnham, and R. J. Colton, *Science*, **248**, 454 (1990).
- ⁸⁸ B. B. Doris and R. I. Hegde, *Applied Physics Letters* (to be published).
- ⁸⁹ A. Zangwill, *Physics at Surfaces*, Cambridge: Cambridge University Press (1988).
- ⁹⁰ The epoxy acrylic coating sample was provided by Dr. J. Li of MMI.
- ⁹¹ The PZT ceramic film sample was provided by Dr. Y. Xu of UCLA.
- ⁹² The sample of MBE grown multilayer was prepared by Dr. Hua Sheng Wu in MSU's CRISS lab.
- ⁹³ S. Gwo, et. al., *Appl. Phys. Lett.* **64**, 493 (1994).
- ⁹⁴ R. Wiesendanger, *Scanning Probe Microscopy and Spectroscopy*, Cambridge: Cambridge University Press, p. 210 (1994).
- ⁹⁵ E. Abbe, *Archiv. Mikroskopische Anat.* **9**, 413 (1873).
- ⁹⁶ E. H. Synge, *Phil. Mag.* **6**, 356 (1928).
- ⁹⁷ M. T. Postek, H. J. Ho, and H. L. Weese, to be published, (1997).

MONTANA STATE UNIVERSITY LIBRARIES



3 1762 10314438 0