2013

Optically Induced Forces In Scanning Probe Microscopy

Dana Kohlgraf-Owens
University of Central Florida

Part of the [Electromagnetics and Photonics Commons](#), and the [Optics Commons](#)
Find similar works at: <https://stars.library.ucf.edu/etd>
University of Central Florida Libraries <http://library.ucf.edu>

This Doctoral Dissertation (Open Access) is brought to you for free and open access by STARS. It has been accepted for inclusion in Electronic Theses and Dissertations, 2004-2019 by an authorized administrator of STARS. For more information, please contact STARS@ucf.edu.

STARS Citation

Kohlgraf-Owens, Dana, "Optically Induced Forces In Scanning Probe Microscopy" (2013). *Electronic Theses and Dissertations, 2004-2019*. 2904.
<https://stars.library.ucf.edu/etd/2904>

OPTICALLY INDUCED FORCES IN SCANNING PROBE MICROSCOPY

by

DANA C. KOHLGRAF-OWENS
B.S., The Ohio State University, 2005
M.S., University of Central Florida, 2007

A dissertation submitted in partial fulfillment of the requirements
for the degree of Doctor of Philosophy
in the College of Optics and Photonics
at the University of Central Florida
Orlando, Florida

Spring Term
2013

Major Professor: Aristide Dogariu

© 2013 Dana C. Kohlgraf-Owens

ABSTRACT

The focus of this dissertation is the study of measuring light not by energy transfer as is done with a standard photodetector such as a photographic film or charged coupled device, but rather by the forces which the light exerts on matter. In this manner we are able to replace or complement standard photodetector-based light detection techniques. One key attribute of force detection is that it permits the measurement of light over a very large range of frequencies including those which are difficult to access with standard photodetectors, such as the far IR and THz.

The dissertation addresses the specific phenomena associated with optically induced force (OIF) detection in the near-field where light can be detected with high spatial resolution close to material interfaces. This is accomplished using a scanning probe microscope (SPM), which has the advantage of already having a sensitive force detector integrated into the system.

The two microscopies we focus on here are atomic force microscopy (AFM) and near-field scanning optical microscopy (NSOM). By detecting surface-induced forces or force gradients applied to a very small size probe (~ 20 nm diameter), AFM measures the force acting on the probe as a function of the tip-sample separation or extracts topography information. Typical NSOM utilizes either a small aperture ($\sim 50-150$ nm diameter) to collect and/or radiate light in a small volume or a small scatterer (~ 20 nm diameter) in order to scatter light in a very small volume. This light is then measured with an avalanche photodiode or a photomultiplier tube.

These two modalities may be combined in order to simultaneously map the local intensity distribution and topography of a sample of interest. A critical assumption made when performing

such a measurement is that the distance regulation, which is based on surface induced forces, and the intensity distribution are independent. In other words, it is assumed that the presence of optical fields does not influence the AFM operation. However, it is well known that light exerts forces on the matter with which it interacts. This light-induced force may affect the atomic *force* microscope tip-sample distance regulation mechanism or, by modifying the tip, it may also indirectly influence the distance between the probe and the surface.

This dissertation will present evidence that the effect of optically induced forces is strong enough to be observed when performing typical NSOM measurements. This effect is first studied on common experimental situations to show where and how these forces manifest themselves. Afterward, several new measurement approaches are demonstrated, which take advantage of this additional information to either complement or replace standard NSOM detection. For example, the force acting on the probe can be detected while simultaneously extracting the tip-sample separation, a measurement characteristic which is typically difficult to obtain. Moreover, the standard field collection with an aperture NSOM and the measurement of optically induced forces can be operated simultaneously. Thus, complementary information about the field intensity and its gradient can be, for the first time, collected with a single probe. Finally, a new scanning probe modality, multi-frequency NSOM (MF-NSOM), will be demonstrated. In this approach, the tuning fork is driven electrically at one frequency to perform a standard tip-sample distance regulation to follow the sample topography and optically driven at another frequency to measure the optically induced force. This novel technique provides a viable alternative to standard NSOM scanning and should be of particular interest in the long wavelength regime, e.g. far IR and THz.

In memory of my grandmother, Patricia Gray Sealey

ACKNOWLEDGMENTS

First and foremost, I gratefully acknowledge my advisor, Dr. Aristide Dogariu, for his advice, support and guidance during my time at CREOL. It has been a privilege and a pleasure to work in his group. I am particularly grateful for opportunity to explore the exciting possibilities to do fun science in the near-field.

I thank the Random group members for the many discussions we had and helpful ideas I received. In particular I would like to thank Dr. Sergey Sukhov for his limitless patience in answering my millions of questions and always helping me find a path away from the circles I would all too often start running. I likewise thank Dr. David Haefner for all of his help and advice regarding the nitty gritty details involved in executing successful near-field experiments. The help and advice about anything and everything I received from Dr. John Broky, Dr. Thomas Kohlgraf-Owens, Kyle Douglass and Lorrene Tripp is also gratefully acknowledged.

For their willingness to freely share and discuss their results on optically induced forces, I thank our French collaborators, Dr. Yannick De Wilde and Léo Greusard. Those discussions and data were instrumental in guiding our work.

I also thank Dr. Yannick De Wilde, Dr. Pieter Kik and Dr. Demetrios Christodoulides for their participation in my dissertation committee as well as for their helpful thoughts and insights. Dr. Pieter Kik was especially instrumental in providing a tremendous degree of advice and instruction. Indeed much of what I know about being an effective graduate student I learned from him.

I likewise thank Dr. Amitabh Ghoshal, Dr. Alex Savchyn, Grady Webb-Wood, and Forrest Ruhge for their boundless willingness to share their time, knowledge and for the many good times we shared both in and out of CREOL.

I was tremendously fortunate to have had many wonderful teachers and professors through the years. There are too many to list here, but one who really stands out in my mind is my high school calculus teacher, Mrs. Karen Austin, who taught me an incredible amount about not only calculus but more importantly about life. She was truly instrumental in positively shaping who I am today.

For sharing all of our duties and responsibilities outside of work and for always meeting me halfway, I thank my husband and colleague, Dr. Thomas Kohlgraf-Owens. Without his help, I could not have finished this dissertation. I also thank my son, Andrew, for being so adaptable particularly these last few months. Watching and hopefully productively assisting your growth and development has been the highlight of my life. I hope you grow to follow your life's path, wherever that may lead, knowing that I love you for who you are.

Finally I would like to thank my grandmother, to whom I dedicate this dissertation, for serving as my hero and my role model. Her keen eye for irony and her subtle and respectful sense of humor along with her unfailing courtesy are sorely missed.

TABLE OF CONTENTS

LIST OF TABLES	xiii
LIST OF SYMBOLS AND ACRONYMS	xiv
1 INTRODUCTION	1
2 SCANNING PROBE MICROSCOPY	7
2.1 Common features of SPMs	8
2.2 Different operation modalities for atomic force microscopy	9
2.2.1 Cantilever vs. tuning fork	9
2.2.2 Contact vs. intermittent contact vs. non-contact	11
2.2.3 Amplitude/phase vs. frequency shift/dissipation feedback	13
2.2.4 Lateral vs. normal scanning	14
2.3 Overview of near-field scanning optical microscopy (NSOM)	15
2.3.1 Aperture NSOM (a-NSOM)	16
2.3.2 Scattering NSOM (s-NSOM)	18
2.3.3 Constant-gap mode (CGM) vs. constant-height mode (CHM)	21
2.4 Artifacts in SPM	22
2.4.1 Artifacts in a-NSOM	22
2.4.2 Artifacts in s-NSOM	23
2.5 The Nanonics MultiView 4000	24
2.6 Forces in SPM	25
2.6.1 van der Waals/Casimir force	25
2.6.2 Other forces in SPM	29
3 OPTICALLY INDUCED EFFECTS ON FORCES IN SPM	32
3.1 Kelvin probe force microscopy	32
3.2 Other direct optically induced force effects	34
3.3 Other indirect (thermal) effects on the cantilever	36
3.4 Other Optically Induced Effects	37
4 ARTIFACTS DUE TO OPTICALLY INDUCED FORCES (OIF)	42
4.1 Forces due to Electromagnetic Fields	42
4.2 Optically induced forces (OIF)	48
4.3 Discussion of the presence of OIF in different modalities	55
5 FORCES IN NEAR FIELD SCANNING OPTICAL MICROSCOPY (NSOM)	58
5.1 Optical forces in propagating fields	58
5.2 Oscillating probe and frequency detuning	61
5.3 Forces acting on probe	62
5.4 Optical force distribution	64
5.5 Optical forces in evanescent fields	68
5.6 Applications	71
6 DISCRIMINATION OF FIELD COMPONENTS IN NEAR-FIELD SCANNING OPTICAL MICROSCOPY	73
6.1 Introduction	73
6.2 Experiment	74
6.3 Simple Probe Model	77

6.4	Conclusion	81
7	MULTIFREQUENCY NEAR-FIELD SCANNING OPTICAL MICROSCOPY (MF-NSOM).....	83
7.1	Introduction.....	83
7.2	Demonstration of MF-NSOM.....	85
7.3	Characteristics of optical force imaging	92
7.4	MF-NSOM Imaging.....	97
7.5	Discussion	101
7.6	Conclusions.....	103
8	SUMMARY OF ORIGINAL CONTRIBUTIONS AND CONCLUSIONS.....	104
	APPENDIX: LIST OF PUBLICATIONS AND PRESENTATIONS.....	108
8.1	Refereed Publications	109
8.2	Contributed Presentations	110
8.3	Contributed Posters.....	112

LIST OF FIGURES

Figure 2-1: Typical cantilever based AFM.....	10
Figure 2-2: Tip mounted in a (a) normal force and (b) shear force mode.	11
Figure 2-3 Common modalities for aperture NSOM measurements: illumination mode collecting (a) reflected and (b) transmitted light, (c) dual mode, collection mode collecting (d) reflected, and (e) transmitted light.	17
Figure 2-4 Schematic of s-NSOM and its model.....	19
Figure 2-5: Picture of MultiView 4000 microscope.	25
Figure 4-1: Schematic of dipole over a surface.	45
Figure 4-2 “Real” surface topography (dash-dotted line) of a Au film deposited on quartz which has been annealed to open up pits in the coating. The solid line is the difference between topographies recorded with and without illumination. The dashed line is the difference between two scans of the real topography performed before and after illumination. Due to optical forces, the tip is pushed ~ 20 nm away at the edge of the Au film.....	51
Figure 4-3 The effect of optically induced forces on a nanostructured GaP sample measured with an uncoated NSOM tip. (a) Optically induced topographies, $\delta z_1 = z_{on} - z_{off1}$ and (b) control difference $\delta z_2 = z_{off2} - z_{off1}$ overlaid on the “real” topography, z_{off1} . Note that the largest artifacts induced by optical forces are located at the edges where the strength of the local z directed intensity gradient is the highest.....	54
Figure 5-1 (Color online) Perceived topography over the core of a single mode optical fiber when (a) 24 and (b) 16 mW of 532 nm laser light is coupled into fiber. The inset in (a) is the side view of the probe; the scale bar corresponds to 100 μm . The inset in (b) shows the measured topography of the fiber face. The blue dashed circles indicate the location of the fiber core which is $\sim 2.2 \mu m$ in diameter. The arrow indicates the orientation of the tip during scanning.	59
Figure 5-2 Gradient of the optical force extracted from the perceived topography shown in Fig. 5-1 when (a) 24 and (b) 16 mW power is coupled into a single mode fiber.	65
Figure 5-3 (a) Simulated probe geometry together with electric field distribution and (b) the calculated (solid line) and measured (symbols) gradient of the optical force acting on a probe scanning over the core of fiber along the direction indicated in the inset of Fig.1b. The error bars account for the errors associated with estimating z and A along with the noise in the topography maps.....	67

Figure 5-4 (a) Topography and (b) extracted optically induced force at the surface of a prism due to a focused laser beam illuminated in a TIR condition.	69
Figure 6-1: Schematic of experiment. An interference pattern is generated on the surface of a prism using two focused laser beams. A standard NSOM tip is scanned over this focused spot to generate a map of the topography for both TE and TM illumination.....	75
Figure 6-2: Topography (force gradient) when (a) TE and (b) TM polarized light is incident on the prism surface; (c) and (d) are the same for the NSOM signal.	76
Figure 6-3: Averaged height of optically induced topography (solid red lines) and NSOM signal (dashed blue lines) for (a) TE polarization and (b) TM polarization.....	79
Figure 6-4: Calculated NSOM signals (dashed blue lines) and gradient of the force signals, which are nonlinearly related to the topography (solid red lines) for (a) TE and (b) TM polarization.	81
Figure 7-1: Schematic of multi-frequency NSOM setup used in the proof-of-concept experiment.	90
Figure 7-2: Magnitude and phase of the current induced on the tuning fork as a result of optically induced excitation (blue solid curves) and electrical driving (red dashed curves). (a) and (b) are the magnitude and phase at the lowest resonance while (c) and (d) are the magnitude and phase at the next higher resonance.....	92
Figure 7-3: Frequency sweep of higher order resonance of uncoated NSOM probe used for scans. Red dashed lines indicate frequencies used for left three columns of Figure 7-4. The blue dash-dot line indicates the frequency used in Figure 7-5.	93
Figure 7-4: Reading down the columns, each shows the topography of the face of a single mode fiber followed by the NSOM signal and amplitude and phase respectively measured at the optical modulation frequency for an uncoated NSOM probe scanning over the fiber's face with 1310 <i>nm</i> light coupled out. The left three columns correspond to frequencies somewhat below ($f = 241.37 \text{ kHz}$), at ($f = 241.59 \text{ kHz}$) and somewhat above ($f = 241.94 \text{ kHz}$) the higher order resonance. The fourth column shows data collected with the light turned off. The rightmost column shows data at an excitation frequency of $f = 241.37 \text{ kHz}$ collected when the probe is many microns above the surface.	95
Figure 7-5: This figure is for the same setup as Figure 7-4 except that the 1310 <i>nm</i> laser diode was replaced with a 635 <i>nm</i> laser diode such that the fiber is now slightly multimode. The resulting topography is shown in (a), the NSOM signal in (b) and the amplitude and phase of the signal from the tuning fork demodulated at the optical modulation frequency are shown in (c) and (d) respectively.....	96

Figure 7-6: Experimental schematic (a) and the measured NSL topography (b) as described in text. Measured amplitude (c) and phase (d) of the tuning fork signal at the optical driving frequency. Color coded amplitude (e) and phase (f) mapped on the 3D topography of NSL. 99

LIST OF TABLES

Table 2-1: Geometrical factors for the determination of the van der Waals force for the interaction of a plane with the following structures in both the nonretarded and retarded interaction regimes.....	28
--	----

LIST OF SYMBOLS AND ACRONYMS

AFM	Atomic Force Microscopy
a-NSOM	Aperture Near-Field Scanning Optical Microscopy
KPFM	Kelvin Probe Force Microscopy
NSOM	Near-Field Scanning Optical Microscopy
OIF	Optically Induced Force
s-NSOM	Scattering Near-Field Scanning Optical Microscopy
SPM	Scanning Probe Microscopy
TE	Transverse Electric
TIR	Total Internal Reflection
TM	Transverse Magnetic
R	Radius
F	Force
θ	Angle
f	Frequency
ω	Angular Frequency
k	Spring Constant
z	Tip-Sample Separation
A	Half the Tip Oscillation Amplitude
t	Time
t_{xx}	Transmission Coefficient

E	Electric Field
ε	Dielectric Constant
α	Polarizability
n	Refractive Index
p	Induced Dipole Moment
β	Wavevector Along Normal Direction

1 INTRODUCTION

In standard imaging, a detector such as photographic film or a charged coupled device (CCD) is used to generate a spatially resolved map of the intensity distribution impinging on the detector. Such detectors rely on the transfer of photon energy to produce a measurable signal. For example, a CCD consists of an array of photodiodes, which rely on the photoelectric effect to convert photons to electron/hole pairs which are used to generate a current proportional to the incident intensity. The bandwidth of photodiodes is limited by the material band gap at the long wavelength limit and prohibitively high absorption at the short wavelength limit [1,2]. Such detectors have many advantages, particularly for detecting light in the visible and near-IR where they are inexpensive, fast, robust, low noise, and operate at room temperature. However the attainable resolution is determined by the size of the pixels or grain size of the photographic film as well as the quality of the optics used to focus the light. Because these systems work in the far-field, such systems at best attain diffraction limited resolution.

To overcome this limit, a number of different techniques have been employed, particularly in the biological sciences such as stochastic optical reconstruction microscopy (STORM), photoactivated localization microscopy (PALM), saturated structured illumination microscopy (SSIM), and stimulated emission depletion (STED) [3–5]. These microscopies rely on preparing fluorescent tags with specific properties which are subsequently attached to the structures of interest. Another possibility is to use a device called a near-field scanning optical microscope (NSOM) which is able to achieve sub-diffraction limited imaging by scanning a very sharp probe in the near-field of a sample where high spatial resolution data is contained. The small probe acts to convert some of the evanescent waves into propagating waves which may be

detected in the far field. Because of the very low light levels involved, typically a photomultiplier tube (PMT) or avalanche photodiode (APD) is used to detect the light. These differ from standard photodetectors used in CCDs by amplifying the signal by inducing a cascade effect whereby one electron may generate many electrons and thus a measureable signal [1,2].

By their nature, these detectors are relatively narrowband. Additionally, the detection of long wavelengths requires a very small band gap so that electrons may be promoted from the valence to the conduction band. As this band gap narrows in the infrared, it becomes increasingly probable for an electron to be promoted by thermal excitation; thus cooling becomes necessary [1,2]. Even more difficult is the realization of sensitive, cost effective detectors in the THz [6].

This argues for the need to detect photons via alternate means. One means is to use so-called thermal detectors, which rely on using a thermally sensitive material to detect light via heat generated by absorption of the photons. Such detectors can be very broadband and have a relatively flat sensitivity, limited by the absorption characteristics of the material. However, their response times are quite long – on the order of milliseconds [7].

Another possibility is to detect light not by energy transfer or absorption, but rather by measuring optically induced forces. This force may have different origins such as radiation pressure or gradients of intensity.

A well calibrated optical trap, as is done in photon force microscopy, may be used to perform force measurements [8,9]. However, another sensitive force detector is a cantilever or tuning fork, particularly when operated on or near its resonance frequency. This is the type of

detection studied in this dissertation. As we discuss in detail, such detectors may be detect light via two mechanisms, first by the direct excitation of induced oscillations and second by the changes in effective resonance properties of an externally driven cantilever when exposed to a force gradient over the probe oscillation cycle. Note that this force gradient is different from the gradient force. The force gradient arises from a gradient in any force, for example a gradient in the radiation pressure or a gradient in the gradient force.

The primary advantage of measuring electromagnetic field distributions via force detection is that a well designed single detector may in principle be used to detect radiation from a large band of frequencies from the THz to the visible and beyond. Indeed such detection is of particular interest in the far-IR and THz regimes where a relative lack of alternatives exists. All that is required is that the photons interact with the probe. Unlike thermal detectors, even a probe which is transparent at the wavelength of interest can be used and, additionally, tuning forks can be dramatically more sensitive to radiation modulated on resonance; in other words they effectively filter noise and off-resonance radiation with higher Q factors corresponding to sharper filters with stronger suppression of off-resonance radiation. To this end, oscillators with Q factors of nearly 10^{10} have been demonstrated with whispering gallery mode resonators [9].

This dissertation focuses on the use of optical force detection to infer properties of light in the near-field. Specifically discussed is the presence and influence of optically induced forces in scanning probe microscopy. We show that these forces can be accurately detected either by examining force gradient induced changes in the resonance conditions of the tuning fork on which the probe is attached or by directly measuring the optical force-induced driving of the tuning fork.

In the practice of AFM and NSOM, a common assumption is that the only additional external force gradients acting on a dielectric probe when it is brought into proximity with a dielectric sample surface are due to the interaction of the probe with the surface, for example from van der Waals/Casimir forces. However, light in addition to emitting photons also exerts a force upon the matter with which it interacts. Even for moderate illumination intensities, these forces are of similar strength to Casimir forces at a tip-sample separation on the order of 20 *nm* away. As we discuss later, such separations are commonly found in standard scanning probe systems. Therefore, as we demonstrate in this dissertation, these forces may significantly contribute to the measured force gradient.

Before we show this demonstration, however, we first provide the necessary background in Chapter 2, where we provide a brief overview of the different options available for atomic force microscope (AFM) and near-field scanning optical microscope (NSOM) systems, previously reported non-optically induced force related artifacts in NSOM measurements, the specific system we use for our measurements along with a description of forces acting in SPM systems with a focus on van der Waals/Casimir forces. In this chapter the description of AFM and NSOM systems is broad in order to encompass as many different systems as possible. We do this because the focus of this research is on optically induced effects on an NSOM; i.e. the NSOM is not a device we use to measure a sample we are studying; it *is* the device under study. Even though we study only one system, the information in Chapter 2 is used to help us build the framework necessary to generalize whether we expect to see similar results for other available systems.

Optical effects on scanning probes can have different origins particularly thermally induced effects, and Chapter 3 provides an overview of previous reports of artifacts present in SPM systems.

Chapter 4 presents experimental evidence for the existence of optically induced forces in NSOM scans. We demonstrate that these effects can induce significant artifacts when measuring optical near-fields; however, once acknowledged they can be accounted for.

The optically induced forces can offer additional measurement possibilities. Next we focus on taking advantage of their inherent presence. In Chapter 5, we demonstrate the quantitative extraction of the optically induced force acting on the probe. In performing this extraction we are able to determine the tip-sample separation at closest approach, a quantity which is often unknown and typically difficult to determine [8,9]. Knowledge of this quantity permits quantitative extraction of the forces acting on the probe as well as more conclusive correspondence between experimental data and theoretical models.

In Chapter 6 we demonstrate that, in the near field, the NSOM and force detection procedures can be complementary. We show that these detection modalities can be operated simultaneously to practically double the amount of collected information without increasing the complexity of the experiment.

Finally, in Chapter 7 we establish the use of force detection as an alternative to NSOM detection. In contrast to the previous chapters where we detected optically induced forces by following optical force gradient induced changes produced in the resonance properties of the tuning fork, here, we demonstrate that a direct excitation of oscillations in the probe is possible and can be induced by modulating the intensity of light interacting with the tuning fork. The

advantages and disadvantages of this direct excitation in comparison with the indirect resonance shift detection are discussed along with a comparison to standard NSOM imaging modalities.

In standard microscopy and imaging, only the field transformations and detector sensitivity must be known in order to characterize the incident intensity distributions. The primary downside of this approach is that the resulting resolution is diffraction limited. NSOM is able to overcome this limit, enabling the measurement of high resolution data. Intrinsic to this measurement is a much richer spectrum of collected information, including the optically induced force effects discussed here. While those interested in using NSOM to obtain high resolution images analogous to standard microscopy may regard the probe's sensitivity to optically induced forces as an artifact to be contended with, we demonstrate that such sensitivity provides, in fact, means to replace or complement standard NSOM measurements, resulting in a wealth of additional information.

2 SCANNING PROBE MICROSCOPY

Scanning probe microscopy (SPM) encompasses a wide range of microscopies which utilize a sharp probe to perform high resolution scans to locally map a desired property on a point by point basis. Specific examples include atomic force microscopy (AFM), near-field scanning optical microscopy (NSOM), magnetic force microscopy (MFM), Kelvin probe force microscopy (KPFM) and electrostatic force microscopy (EFM). Each of these microscopies measures a different physical quantity with a resolution which depends on the specific probe being utilized, the sample under study and a host of environmental considerations such as whether the scan is performed in vacuum or ambient conditions. We discuss a representative sample of these in section 2.6. While many SPMs exist, the focus of this work is AFM and NSOM.

Standard AFM and NSOM probes are fabricated using techniques such as pulling optical fibers, chemical etching, and nanofabrication [10,11]. Applying a metal coating at an oblique angle with respect to the tip allows us to confine light within the tip while keeping the aperture open, creating aperture NSOM (a-NSOM) probes [10]. Scattering NSOM (s-NSOM) typically utilizes metal or metal coated AFM probes in order measure a relatively strong near-field light signal over a very small volume. AFM scans are typically performed with uncoated probes in order to maximize the attainable resolution.

The variety of signals which may be measured locally using scanning probe microscopes is limited only to the imagination and technical prowess of the probe manufacturer. Depending on how the probes are fabricated, they may be used to detect a host of physical quantities at the nanoscale. Limiting oneself to pulled nanopipette probes, a standard one may be used for

nanoscale delivery of glasses or liquids, housing fluorophores in a small volume or delivering high power laser light. A metal wire may be added to the pipette to create an electrochemical sensor; then by coating the outside of the pipette with metal, a nanoscale thermocouple or coaxial cable can be realized. Using a dual channel pipette, two electrically isolated wires can be pulled and used either to measure thermal conductivity, capacitance or resistance on the nanoscale or alternatively as nanoscale tweezers [12]. On a lighter note, the cantilever of a SPM probe can also be used as a base for mounting micro-scale figurines [13].

Different operating modalities have been developed for AFM and NSOM to explore a host of physical phenomena. In this chapter, we provide an introduction to many of the options available. Good overviews of many of the topics discussed in this chapter may be found in, e.g. [10,14–16]. *Because SPMs are by their nature interaction microscopies, the exact configuration of the microscope is critical in determining what precisely is measured.*

2.1 Common features of SPMs

Scanning probe microscopies sacrifice the ability to collect data in parallel in order to achieve higher spatial resolution by sampling the physical quantity of interest point by point with a sharp probe. They are by their nature interaction microscopies; thus determination of what they measure must be done in a self-consistent manner. These microscopies have opened up the possibility to explore numerous phenomena on more localized scales than otherwise would be possible at the expense that interpreting the collected data is relatively challenging. Indeed, significant attention has been devoted to understanding exactly what these microscopes measure as well as the artifacts which might skew the correct interpretation of the collected data.

In the rest of the section we delve into the details of how representative samples of SPMs differ. In essence the difference lies in the precise physical phenomena which act as the predominant contributor(s) in the interaction between the tip and the sample. However in all cases the fundamental mechanics of the measurement remain the same.

2.2 Different operation modalities for atomic force microscopy

Atomic force microscopy (AFM) is a technique which is used for the nanoscale detection of forces. Typical force sensitivity is on the order of piconewtons, but probes with sensitivity at least down to the attonewton force scale have been reported [17]. Using this device, it is possible to image samples even down to the single atom level [18].

This section provides an overview of the many different options available in commercial AFMs. As with the rest of this chapter, this section is not meant to be exhaustive but rather is intended to provide an overview of important types of AFM devices.

2.2.1 *Cantilever vs. tuning fork*

The first major option at the user's disposal is whether the probe is mounted to a tuning fork or a cantilever. These two options provide complementary methods to sensitively detect external forces and thus provide the vital sensing necessary in the operation of an atomic *force* microscope.

The first option is a cantilever based system. A typically experimental setup is shown in Figure 2-1. Here a probe with a small aperture ($d \sim 20 \text{ nm}$) is mounted to the end of a cantilever. Information about the surface topography is provided by a laser which bounces off of the back of this cantilever and onto a two or four section photodiode diode, where a two section

diode will determine the vertical deflection of the cantilever and a four section photodiode will also detect friction via the torsion of the probe [19]. This cantilever may either be operated in static mode or electrically driven near its resonance frequency and operated in either non-contact or intermittent contact mode.

Figure 2-1: Typical cantilever based AFM.

Alternatively, the probe can be attached to one of the arms of a tuning fork which is electrically driven near its resonance frequency. This system operates very similarly to the cantilever based system except that the feedback is entirely electrical. Depending on whether there is a bend in the fiber or not, the tuning fork may be either operated in normal force or shear force mode. With normal force the tuning fork is more or less parallel to the sample, shown in Figure 2-2(a), so that the tip oscillates predominately perpendicular to the sample; for shear force, shown in Figure 2-2(b), the tuning fork oscillates predominantly parallel to the sample. Because this device must be electrically driven, it is only able to operate in intermittent contact

or non-contact mode. Details about the signals which may be measured are discussed in more detail in section 2.2.3..

Figure 2-2: Tip mounted in a (a) normal force and (b) shear force mode.

2.2.2 *Contact vs. intermittent contact vs. non-contact*

The next option is what kind of contact the probe makes with the sample. Three primary options exist: contact, non-contact and intermittent contact mode [20]. A cantilever may either be electrically driven in intermittent contact or non-contact mode or remain static in contact mode, but a tuning fork must be electrically driven in either intermittent contact or non-contact.

In contact mode a tip is brought in physical contact with the surface and then dragged along the sample during the course of a scan. The cantilever responds to changes in the topography, i.e. changes in the force applied to the cantilever, by deflecting. Typically feedback is used to maintain a constant deflection on the cantilever thus keeping the tip sample separation constant, thereby permitting one to measure a map of the local topography. Alternatively, the feedback may be turned off and the deflection measured. Because the excursions of the cantilever during a scan are small, the externally applied force is relatively straightforward to

determine. This is a huge advantage of contact mode scans. However because the tip is physically brought into contact and remains in contact for the duration of the scan, this type of scan is relatively abusive to both the tip and sample. Therefore this modality is particularly ill-suited to scan very soft samples such as biological samples. In addition, an appropriate filter must be added before the NSOM signal photodetector in order to block stray light from the beam bounce laser.

Alternatively, one can scan in non-contact mode. In this modality, a tip is oscillated near its resonance frequency with very small oscillation amplitude (typically less than 10 *nm*). This tip is then brought close to the sample surface without coming into contact with it. The tip-sample separation may be regulated based on changes in the amplitude or phase of cantilever at the driving frequency or the frequency shift/dissipation in the cantilever, as described in the next section. Because contact is not made, neither the probe nor the sample is worn or damaged due to the other. However, in ambient conditions, a thin layer of water forms on a surface, which, in non-contact mode, affects the measured topography. Therefore this type of scan is typically only performed in vacuum.

A compromise between these two options is a technique called intermittent contact mode. This modality operates similarly to non-contact mode except that the tip oscillation amplitude is much higher, typically tens of nanometers. Because of this much high oscillation amplitude, the restoring force is strong enough that the tip is able to come in physical contact with the sample and then overcome the strong attractive forces close to the sample and pull away. Thus the tip is able to penetrate the surface water layer and image the sample underneath. Because such a

technique is less damaging to the probe and sample as compared with contact mode AFM and yet is able to image the sample in ambient conditions, it is a popular imaging modality.

2.2.3 *Amplitude/phase vs. frequency shift/dissipation feedback*

For oscillating probes, several different signals may be utilized to collect data about the system properties or provide feedback control. For driven cantilevers, either amplitude or phase signals may be utilized. Tuning fork based systems may also utilize the complementary frequency shift or dissipation signals. Additional information about a sample may be gleaned by either reading both signals or maintaining a feedback loop based on one and reading the other. Here we discuss utilizing these signals for feedback purposes.

For amplitude or phase feedback, the tuning fork is electrically driven near the tuning fork resonance frequency. When the probe is far away from the sample, the probe oscillates with an amplitude A_0 and a phase ϕ_0 relative to the driving force. When the probe gets close to the sample or another additional external force is applied to it, the resonance frequency of the probe shifts due to the presence of force gradients over the probe oscillation cycle. Damping may also be introduced if, for example, the probe contacts the sample surface and transfers energy to it or is immersed in a liquid such as water. These changes will affect the amplitude and phase of the oscillation on the probe, which are measured via a lock-in. By maintaining a constant change in the amplitude or phase at the tuning fork driving frequency, a feedback loop may be applied to control the tip-sample distance. An alternative approach for systems based on tuning forks is to measure the frequency shift or dissipation on the probe directly and to control the tip-sample

distance by maintaining a constant frequency shift or a constant level of dissipation on the probe [21–23].

One may ask why for oscillating probes, we measure the gradient of the force rather than the force itself. The answer lies in the fact that for oscillating probes, the quantities we measure are related either directly or indirectly to changes in the properties of the tuning fork, i.e. a shift in the resonance frequency or a change in oscillation amplitude at the driving frequency. In the simplest case when the gradient of the force is constant over the entire oscillation amplitude, the change in the resonance frequency, Δf , is given by $\Delta f = (f_0/2k_{sc})(-\partial F_{ts}/\partial z)$ where f_0 is the resonance frequency, k_{sc} is the spring constant and $\partial F_{ts}/\partial z$ is the gradient of the tip sample force in the direction normal to the sample surface. This assumption often works well for non-contact mode SPM where the tip oscillation amplitude is very small. For intermittent contact mode, a more general solution for the case where the force gradient varies significantly over an oscillation cycle has been derived in for example Ref. [18].

2.2.4 *Lateral vs. normal scanning*

Another option at our disposal is whether to scan vertically or laterally with respect to the sample. Of course, the piezo controlling the scanning of the tip and/or sample is capable of moving in all three directions. However volume scans are typically not performed because of their large time requirements and increased difficulty. When feedback is engaged, lateral scans allow the operator to closely follow the surface topography of a sample, enabling high resolution mapping of the 3D surface profile as well as the manipulation of evanescent optical fields in the near zone of the sample. Alternatively one can perform force-distance curves by scanning the

probe vertically at specified locations along the sample. This type of scan also goes by the name force spectroscopy [20].

2.3 Overview of near-field scanning optical microscopy (NSOM)

When considering the options available for NSOM scanning, the first and most obvious option is between scattering NSOM (s-NSOM) and aperture NSOM (a-NSOM). Before we detail the differences between a-NSOM and s-NSOM, we will briefly review what they have in common. At their essence, these imaging modalities utilize a sub-wavelength tip or aperture placed very close to the sample which acts to convert propagating to evanescent and/or evanescent to propagating electromagnetic radiation. Such a conversion is necessary because evanescent fields, which permit us to break the diffraction limit of light, decay exponentially and thus are typically not detected. The precise involvement of the probe in this process depends on the particular imaging modality employed, as we will detail below.

Like other SPMs, NSOMs are interaction based microscopes. Indeed the interaction of the probe with the complex 3-D field distribution that it generates and/or samples remains a topic of study even today. It depends on many parameters, including the exact geometry of the probe, which is at a minimum difficult to characterize and tends to change over the course of its lifetime due to subtle and not so subtle damage it endures. It also depends on a host of physical processes on the sample and probe with which the light interacts in expected and all too often unexpected ways. Some of these dependencies are discussed in detail in section 2.4 and chapter 4.

Ideally this probe does not significantly perturb the very radiation it is measuring. Dielectric probes obviously modify the radiation less than metal coated probes, but typically these measurements are performed with a metal or metal coated probe in the case of s-NSOM in

order to increase the scattered signal to be detected or a metal coated probe in a-NSOM in order to achieve sub-diffraction limited resolution. The presence of these metals significantly alters the field distribution, and thus the complex 3D field distribution should be solved self-consistently. In addition the incident radiation may modify the physical processes or properties of the sample or probe. One well-known example of this is absorption induced heating of the metal at the tip of metal coated probes which can lead to lattice expansion and consequently effects such as tip shortening or elongation and changes in metal work function [24–28].

2.3.1 Aperture NSOM (a-NSOM)

Aperture NSOM (a-NSOM) utilizes a probe with a sub-wavelength aperture to illuminate and/or collect radiation above the sample. These probes may be manufactured in a variety of manners. The first technique is to locally heat and pull a fiber or capillary until it breaks [10]. The taper angle and final aperture size may be somewhat controllably produced by controlling the exact pulling parameters. Typical aperture sizes are of the order of 50 – 150 *nm*. Although smaller apertures may be formed with this method, the light coupling efficiency is prohibitively low and thus the aperture size is practically limited to $\sim \lambda/10$ or about 50 *nm* in the visible [29]. In addition to heating and pulling the fibers, they may be chemically etched to realize a variety of probe apertures [10,11]. Another method is to fabricate the probes using Si processing techniques [10].

Several options exist to excite the sample and collect the resulting radiation distribution. These are summarized in Figure 2-3 below.

Figure 2-3 Common modalities for aperture NSOM measurements: illumination mode collecting (a) reflected and (b) transmitted light, (c) dual mode, collection mode collecting (d) reflected, and (e) transmitted light.

These modalities may be grouped into three categories: illumination mode, dual mode and collection mode. In illumination mode (Figure 2-3(a-b)) the excitation is provided through the probe which converts propagating fields to evanescent fields. This light then interacts with the sample, which converts some of the evanescent light into propagating light to be detected either in reflection or transmission mode. Dual mode (Figure 2-3(c)) utilizes the probe to both excite the sample with evanescent fields and then collect the scattered evanescent radiation and convert it to propagating modes to be detected. Finally collection mode NSOM (Figure 2-3(c-d)) selectively collects evanescent fields close to a sample surface and converts them to propagating waves to be detected in the far field. The incident radiation in this case may either be of propagating fields or evanescent fields often excited via total internal reflection (TIR). This discussion assumes a small aperture metal coated NSOM probe is used. If the aperture is not sufficiently small or is not coated with metal, it will also efficiently collect and/or radiate propagating fields.

The aperture of a metal coated probe may be modeled as the combination of an in-plane electric and in-plane magnetic dipole moment whose orientation and strength are specific to the particular tip being utilized [30]. The in-plane magnetic dipole originates as the solution for the transmission of light through a sub-wavelength aperture in an infinite perfect metal conductor for normally incident light [30–32]. The in-plane electric field originates from the deviations of a real NSOM tip from this model, including the fact that it is composed of a tapered optical fiber and the coating is of a real metal deposited with a finite thickness and roughness.

It should be noted that measurements taken in illumination and dual modes are intrinsically interferometric, especially for metal coated probes. In illumination mode, this interference arises due to the multiple reflections of the light between the tip and the sample in illumination mode [33,34]. Similarly in dual mode, the interference predominately arises due to the standing wave that forms between the incident and collected radiation [35]. In principle, collection mode measurements should not be intrinsically interferometric if a small aperture is used. This is because such a small aperture will only effectively couple near-field radiation, thereby effectively eliminating a background signal for it to interfere with. We mention this intrinsic interference term here because it can be a significant contributor to a-NSOM imaging artifacts, which we discuss in detail in section 2.4.

2.3.2 *Scattering NSOM (s-NSOM)*

In the case of s-NSOM, a very sharp probe ($d \sim 20 \text{ nm}$), which is typically metal or metal coated, is oscillated near the cantilever resonance frequency in the near zone of a sample. These probes are either substantially similar to or the same ones used for AFM imaging. The achievable resolution is approximately given by the radius of the probe [36], which is

comparable to the resolution achieved when performing AFM scans and significantly better than what can be achieved using a-NSOM. In addition, these probes do not require thick metal coatings like a-NSOM probes do, permitting the simultaneous measurement of topography and light signals with very fine resolution. Also unlike a-NSOM, the practical size of the aperture does not depend as strongly on the wavelength. Thus the same 20 *nm* diameter probe may be used to image the fields in, for example, the visible and the IR thus allowing for remarkable resolution as a function of wavelength at longer wavelengths.

The tip used in s-NSOM may be modeled as a dipole polarized perpendicular to the sample surface; thus the measured near-field signal may be modeled as the sum of this dipole and its image dipole. This is schematically shown in Figure 2-4. Because the s-NSOM near-field signal predominately arises due to a perpendicularly oriented dipole, it is of crucial importance to excite the sample with that polarization state of light in order to maximize the collected near-field signal [36].

Figure 2-4 Schematic of s-NSOM and its model.

Compared to a-NSOM, where there is a relative plethora of imaging modalities, s-NSOM experiments are carried out in basically the same manner. The tip and sample are externally illuminated with a focused laser beam. A detector is placed at some angle in the far field and is used to collect the scattered radiation. Most of this radiation is background signal from the reflection and scattering of light from the sample as well as reflections off the probe. On top of this very large background is the desired near-field signal. This signal originates from the field scattered by the tip of the probe. Because evanescent fields are generated by both the probe and the sample, this signal depends strongly on the tip sample separation. The signal is rather weak and it is usually extracted by using a lock-in detection at or at a harmonic of the probe oscillation frequency. While scattering from the sample may also convert near field into far field radiation, it is not affected by the probe oscillation and, therefore, is registered as part of the background. In fact, the strength of the scattered signal varies so strongly on the tip-sample separation that further background suppression is routinely achieved by locking-in on a harmonic of the oscillation frequency [36]

Similar to the illumination and dual mode modalities in a-NSOM, s-NSOM is intrinsically interferometric when the signal is collected at the fundamental lock-in frequency [36]. In this case, the interference arises due to the fact that light reflecting off the tip and other background sources which also oscillate with the probe adds coherently with the desired near-field signal. However, this may be dramatically suppressed by locking in on a higher harmonic of the tip oscillation as long as one is careful to ensure that the tip does not press too far into the sample during these data collects. Otherwise, significant higher order contributions to the tip oscillation will also be present, thus introducing significant contributions

of this background signal to the higher harmonic signals as well [36]. These artifacts are discussed in more detail in section 2.4.

2.3.3 *Constant-gap mode (CGM) vs. constant-height mode (CHM)*

Two predominate scanning modalities exist for performing lateral scans with oscillating probes. The first is constant gap mode (CGM), wherein the tip-sample separation is ideally kept constant by utilizing force gradient detection based feedback. The advantage of this is we can ideally maintain a constant near-field resolution in our scans. This is particularly imperative for s-NSOM where, by locking in on the typically second harmonic, the collected signal is typically confined to only the first several *nm* above the surface. Thus one must maintain close contact with the sample in order to detect this signal. The disadvantage of this modality, however, is the presence of artifacts which frequently distort or even overwhelm the desired near-field signal. This is discussed in detail in section 2.4.

Alternatively, constant height mode (CHM) scans are performed by turning off the feedback and scanning the tip at a constant height above the sample. The advantage is we can largely avoid the artifacts which plague CGM scans, but this comes at a large price: namely the variable measured resolution as well as the relatively high experimental difficulty of closely following a sample without crashing into it. Therefore, the utility of this modality is practically reserved for small area scans of relatively flat samples.

A third, much less common modality exists called constant intensity mode (CIM) [10]. This modality works by following the contour of constant intensity and has been shown to be equivalent to CHM when the maximum height of the topography is much smaller than the wavelength of light [37,38].

In principle both a-NSOM and s-NSOM scans may be performed in either CGM or CHM. In practice, s-NSOM scans are almost always done in CGM, and it is often employed for a-NSOM scans as well. For this reason, an understanding of the artifacts present in this imaging modality is of crucial importance.

2.4 Artifacts in SPM

2.4.1 *Artifacts in a-NSOM*

As we have stated many times already, scanning probe microscopies are interaction microscopies. Therefore to properly identify sample properties, the user ideally must minimize, account for or decouple these interactions where possible. Implementing these requirements, however, is usually non-trivial and, in many instances, even impractical. It is nonetheless important to be aware of the interactions that occur that can affect the measurement, if only to qualitatively assist in data interpretation and to guide the data collection. To this end, several groups have undertaken theoretical and experimental studies to attempt to understand this interaction. Here we focus on reported imaging artifacts that can occur when performing NSOM scans.

The predominant artifact that has been discussed is the so called z-motion artifact, which was reported in an experimental paper in 1997 [37] and quickly followed up by a theoretical paper discussing the issue [39]. This artifact is due to the z-motion dependence of the collected optical signal. Indeed, significant variations in the measured NSOM intensity may be seen even when retracting the tip merely 0.1 nm over an unstructured glass surface [37]. It is also

sufficient to allow one to clearly see an index variation between the core and cladding of a standard optical fiber [40].

Unfortunately no good method exists to account for this artifact. It can be prevented by turning the feedback off and scanning at a constant height (CHM) rather than at a constant tip sample separation (CGM) [37], which is typically impractical. Even if a CHM mode scan is performed, a topographic scan should still be separately performed in order to identify the topographical contributions to the signal [41], which significantly adds to the data collection demands. This artifact may be corrected by collecting data not in a 2D plane but rather in a 3D volume above the surface. In doing so, it is possible to separate out signal variations from the z motion of the tip and due to refractive index variations of the sample [35,42]. However, this is also generally not practical due to the difficulty and significant time requirements necessary to collect data in a 3D volume. This leaves us with the question of how to tell if the artifact is significantly present in an image. We can say its effect is small if the topographic and NSOM images are highly uncorrelated globally and locally, a constant displacement exists between correlated structures, or the two images exhibit a different resolution [37].

2.4.2 *Artifacts in s-NSOM*

Similar artifacts are also present in s-NSOM. Indeed, NSOM signals collected at the fundamental frequency of the tip oscillation contain a strong background signal. However, it is possible to lock-in on higher harmonics of the scattered signal. This significantly reduces the background signal at the expense that it also significantly reduces the desired near-field signal [41,43,44]. A good compromise is to lock-in on the second harmonic of the tip oscillation amplitude, in which case the background is strongly suppressed but the optical signal is still

strong enough to be measured. In this instance, strong background suppression is obtained because the second harmonic is sensitive to the gradient of the gradient of the field over the range of tip oscillation, which is only significant for the near fields [41]. A more rigorous description of this effect is given in [36]. However, it should be pointed out that the specific tip modulation amplitude has a significant effect on what field components are preserved and thus the fidelity of the final image. Because the 2nd harmonic will only reflect gradients of the field over distances of the order of the tip modulation amplitude, if a small amplitude is used, then even the lower spatial frequency near-fields will be effectively filtered, leading to strong edge enhancement and the perception of higher resolution. Therefore, the true near-field image is better preserved using a larger tip modulation amplitude [44].

2.5 The Nanonics MultiView 4000

For all experiments included in this dissertation, we used a Nanonics MultiView 4000 scanning probe microscope, the microscope portion of which is shown in Figure 2-5. This microscope utilizes pulled fiber probes glued to a tuning fork operating in normal force mode offering the options of amplitude or phase feedback.

Figure 2-5: Picture of MultiView 4000 microscope.

2.6 Forces in SPM

Now that we have discussed some of the different configurations available for AFM and NSOM, we briefly look at the different forces acting on SPM probes. Because our scans typically involve dielectric samples scanned with uncoated or metal coated dielectric probes, the forces involved in these systems will constitute the bulk of the discussion in this section.

2.6.1 *van der Waals/Casimir force*

One force present in all CGM scans is the van der Waals/Casimir force which is due to the electromagnetic interaction of fluctuating dipoles of the atoms in the sample and the tip. This

force is always present; however, being one of the weakest forces, it is often overwhelmed by other competing forces. The van der Waals interaction potential is generally composed of three parts, each proportional to $1/r^6$ for two interacting dipoles. For electrically neutral atoms, such dipoles are created via the fluctuations of the electron charge density which then induces dipoles in other atoms. The interaction of these dipoles generates the so-called dispersion or London force. Polar molecules, by contrast, have permanent dipoles which can induce dipoles in other atoms generating the so called induction or Debye force. The interaction of these permanent dipoles with each other is orientation dependent, the result of which is the so called orientation or Keesom force. These last two together give the polar force [15,45].

Below we list some general features of the dispersive (polar) van der Waals force between two dipoles/atoms/molecules.

1. This force is typically attractive, although it can be negative when the refractive index (dielectric constant) of the medium between the two dipoles has a refractive index (dielectric constant) between that of the two dipoles. In addition, it is always attractive when the tip and sample are composed of the same materials.
2. It is greatly reduced if the intervening material is not air or vacuum.
3. It is anisotropic, just as the polarizabilities of most molecules are anisotropic.
4. It is non-additive because neighboring molecules interact with one another.
5. The dispersion term of the van der Waals force suffers from retardation effects.

This occurs because at large enough separation, the time it takes for field from the dipoles in one structure to reach the other and come back becomes comparable to the time over which the dipoles fluctuate. This results in the dispersion term

approaching $1/r^7$ rather than being proportional to $1/r^6$. Consequently three interaction regimes exist. For very close separations, the interaction energy between two dipoles is proportional to $1/r^6$. Then for intermediate separations where the retarded dispersion term dominates, it is proportional to $1/r^7$. This occurs at distances of about 100 nm but starts coming into play at much closer separations. Finally the polar term, which is not affected by retardation, will dominate if present and the interaction energy is again proportional to $1/r^6$ [15].

The total force from all of the atomic interactions between the tip and sample contribute to the total force, which is typically on the order of piconewtons to nanonewtons. The actual force depends not only on the material properties of the tip, sample and intervening material, but also on the geometry of the two. If we model the sample as a plane, then the van der Waals force is given by

$$F = \frac{-g_{n,r} A_{n,r}}{D^{n_{n,r}}} \quad (2.1)$$

where g and n are factors listed in Table 2-1 for different probe geometries, A is the material specific and geometry independent Hamaker constant, and D is the tip-sample separation at the point of closest approach. The subscripts n and r stand for non-retarded and retarded respectively [15,46].

Table 2-1: Geometrical factors for the determination of the van der Waals force for the interaction of a plane with the following structures in both the nonretarded and retarded interaction regimes.

	$\mathcal{G}_n$	$\mathcal{G}_r$	n_n	n_r
Cone	$\tan^2(\theta)/6$	$(\pi \tan^2(\theta))/3$	1	2
Sphere	$R/6$	$(2\pi R)/3$	2	3
Paraboloid	$I_{xy}^2/(12I_z)$	$(\pi I_{xy}^2)/(3I_z)$	2	3
Cylinder	$R^2/6$	πR^2	3	4
Plane (Force/unit area)	$1/(6\pi)$	1	3	4

Here θ is the semiaperture of the cone, R is the radius of the sphere or cylinder, I_{xy} and I_z are the semiaxes of the paraboloid. In addition, the sphere tip model in the non-retarded case may be corrected to take into account the elongated nature of the probe using the following equation:

$$F = \frac{-A_n R}{6D^2} \left(1 + \frac{D \tan^2(\theta/2)}{R} \right) \quad (2.2)$$

for a probe of semiaperture $\theta/2$, and length L with $D \ll L$. When $D \ll R$, this correction factor is insignificant [15].

We assume the van der Waals/Casimir force is the dominant surface force present in our scans. As a final note, in the section title we referred to this as the van der Waals/Casimir force. We use both terms due to the seeming lack of concrete definition of what distinguishes them [47,48]; indeed they are even used interchangeably [49].

2.6.2 Other forces in SPM

In general, forces of other origins may also be locally measured using a sharp probe. Similar SPM techniques exist to measure these forces; indeed the largest difference is arguably their name. To first order, what will be measured is the predominant force that interacts with the probe. This predominant force is determined by the particular sample, probe, surrounding environment and exact scanning technique (i.e. non-contact vs. intermittent contact). Here we list a few common forces as well as the names of the techniques used to measure them.

The first we mention is the *image force*, which, as the name implies, is the force due to the formation of image dipoles which is due to the interaction of isolated charges with conducting materials [45]. Image forces encapsulate the many forces that can arise due to the presence of inhomogeneous charge distributions. This includes capacitance forces which we discuss in some detail next, electrostatic forces which we discuss after that and double layer forces which are due to the counterions that form above the surface of a charged liquid layer [15].

A *capacitance force* may also be generated if either a difference exists between the work functions of the tip and sample or there is an applied *electrostatic force* between two conducting or semiconducting materials that are brought into sufficiently close proximity to allow charges to flow between the two surfaces. Of course, an applied electrostatic force is usually applied to cancel the work function difference, but the fidelity of this cancelation on the relevant SPM length scales is not perfect because of the spatial variation of the work function on the nanoscale due to the inhomogeneous charge density distributions that result from the local surface inhomogeneities (i.e. roughness and impurities) of any real surface. This force can dominate all

other forces even for separations as small as several nanometers, making it difficult to avoid for intermittent contact and non-contact mode SPMs. Many scanning probe techniques have been developed to measure the capacitance force. For example, Scanning Spreading Resistance Microscopy (SSRM) and Scanning Capacitance Microscopy (SCM) were developed to map the dopant density distributions in semiconductors via measurements of the local resistivity and capacitance respectively. Additionally, Kelvin probe force microscopy (KPFM), also known as Scanning Surface Potential Microscopy (SSPM), is used to map the variation of the work function across a surface [50]. This technique and the effect light has on it is discussed in more detail in section 3.1. We note that this force is accounted for when calculating the image force so long as the calculation takes into account the effect of an applied bias [45].

Forces can also arise due to the local charging of insulating samples and tips where the charge cannot conduct away. This results first in charge-charge forces, which for a neutral source are short range and thus are typically unimportant. However, the presence of these charges tends to cause atoms to deviate from their expected location in the lattice, causing the formation of dipoles, which result in charge-dipole and dipole-dipole interactions that can introduce an *electrostatic force* which is longer range and thus more significant. Additionally, extra charge in a conducting material can increase the strength of the image force [45]. These electrostatic forces may be measured using Electrostatic Force Microscopy (EFM) [50].

Magnetic forces, by which we mean magnetostatic forces, contribute significantly if both the sample and probe are ferromagnetic. The measurement of such forces constitutes the basis of magnetic force microscopy (MFM) and is typically realized by attaching a ferromagnetic sphere to the tip of a probe and using it to scan a sample in a manner similar to an AFM scan [45,50].

Capillary (meniscus) forces are also present unless an experiment is done in ultra high vacuum and are due to the effects of a thin layer of liquid that forms on top of a sample. When the tip reaches sufficiently close to the sample, it causes the tip to “jump into contact” as a meniscus forms between the tip and sample. This layer will then compress until the tip makes “hard contact”. Upon retracting the probe, this layer stretches until it finally breaks and releases the tip [15,45]. A major side effect of this force is that non-contact mode AFM performed in ambient tends to image this liquid layer rather than the sample beneath. These forces may be evaluated by measuring force-displacement curves [15,45].

Forces may also be present due to *electron transitions* that can occur when a conducting probe is brought into very close proximity (about 0.5 nm separation between the lead atom on the tip and sample) of a conducting sample, such that they affect each other’s electron structure and electron dynamics. This leads to the transport of electrons from one side to the other and thus changes in the system energy [45].

3 OPTICALLY INDUCED EFFECTS ON FORCES IN SPM

Scanning probe microscopes have been used to probe a variety of surface and subsurface properties because of their high sensitivity and high spatial resolution. For instance, atomic force microscopy (AFM) is used to measure forces on the order of piconewtons or less with nanometric resolution. As discussed in Chapter 2, typically, a small probe is scanned either vertically to generate force-distance curves or laterally to map a desired physical quantity. The intensity of a light field can be measured with a near-field scanning optical microscope (NSOM) either by collecting light with a sub-wavelength aperture (a-NSOM) or by collecting the scattering off of a sub-wavelength probe (s-NSOM). The use of scanning probe microscopes to measure external forces such as van der Waals, meniscus, electrostatic, magnetic etc. has been extensively documented in the literature [15,51]. Therefore with the intense study of the different forces which may be sensitively measured in SPM systems coupled with the frequent measurement of light, which is well known to exert a force upon matter, one must ask whether this effect of optically induced forces on scanning probe microscopes has been discovered. The answer is naturally yes. Here we provide a discussion of the previously reported literature in the area.

3.1 Kelvin probe force microscopy

The original and arguably most thoroughly studied microscopy to look at the effect of light is the so called Kelvin probe force microscopy (KPFM) technique. This technique relies on the measurement of local variations in the contact potential difference (CPD) and was experimentally demonstrated in 1991 [52,53] although these demonstrations did not examine the

effect of light on the CPD. The contact potential difference between two conductors depends on several factors, including the material work functions, surface impurities, oxide layers, humidity, dopant concentration for semiconductors and temperature. For bulk materials this difference may be measured using the Kelvin technique wherein the two materials are brought together in close proximity in a parallel plate capacitor arrangement. In this case, the CPD for an ideal parallel plate capacitor arrangement is given by $V_{CPD} = q(\Phi_2 - \Phi_1)$ where V_{CPD} is the voltage due to the CPD, q is the electron charge and Φ_x is the work function of material x including all modifications due to impurities, oxide layers, etc. Oscillating the plate separation at frequency ω generates a current $i(t)$ given by $i(t) = V_{CPD} \omega \Delta C \cos(\omega t)$ where ΔC is the change in capacitance. The contact potential voltage is then measured by applying an external voltage which cancels V_{CPD} and thus drives the current to zero [52].

In order to measure these properties locally, one of the plates is replaced by an AFM tip and the applied force rather than the induced current as a function of applied external voltage between the tip and sample is measured. This force is given locally by $F = -0.5V_{CPD}^2 (\partial C / \partial z)$ where C is the capacitance in the tip-sample junction and z is the tip-sample separation [53]. In order to simultaneously acquire topography and CPD maps, the external voltage and the cantilever oscillation may be driven at different frequencies and separately locked-in on [52].

When light illuminates this junction, the resulting applied optically induced force may be directly measured. Additionally, because conductors and/or semiconductors are involved, this radiation may be absorbed causing thermally induced effects on the tip and/or sample which indirectly affect the CPD between the tip and sample and can therefore be measured by the

Kelvin force probe microscopy technique. An example would be a change in the material work function due to thermally induced lattice expansion of the material. In this manner, local changes in the optical absorption of the tip-sample system may be measured. This was demonstrated in 1992 where the effect of temperature as well as light wavelength was explored [28]. These measurements are typically performed between two conductors or semiconductors, although the sample is not required to be conducting; in this case only the contact potential of the probe varies, giving rise to measurements of the local field intensity [28]. After its initial demonstration, this technique was explored in great detail [54–57]. However, this technique suffers from the requirement that at least the probe must be a metal or a semiconductor. This includes the possibility to use metal-coated probes for Kelvin probe force microscopy imaging [58].

Closely related to this, in 1991 W. Denk and D. W. Pohl reported illumination induced changes in the measured damping of metal coated Si tips over GaAs samples which were sensitive to the illumination strength, sample conductivity and voltage between the tip and sample. They attributed this to the resistive loss of influence currents [59,60].

3.2 Other direct optically induced force effects

To our knowledge, the prospect that an atomic force microscope may be sensitive to direct optically induced forces was first put forth in a theoretical article in 1992 [61], wherein the authors calculated the force on a sphere in an evanescent field from a TIR wave on a triangular prism. Based on their calculations, they concluded that for reasonable excitation intensities, a force should be measurable using a standard AFM. This was then followed up two years later with two more theoretical papers which further refined the idea [62,63].

Since then several papers, both theoretical and experimental, have appeared demonstrating the direct or indirect effects of light on the probe-sample interaction. A direct effect is for example due to optically induced forces which compete with van der Waals/Casimir forces to alter the surface force profile that the AFM probe follows and acts to “trick” the probe into thinking topography exists when it doesn’t, an effect we refer to as the “*topography of light*”. Indirect consequences may be due to thermal heating which can induce lattice expansion in the sample or tip, in the latter case leading to so called tip elongation. These effects are similar to phenomena affecting the Kelvin probe discussed in detail above with the main difference, however, that in KPFM the predominant non-optically induced force is electrostatic in nature.

In terms of theoretical studies of optically induced effects in NSOM as opposed to KPFM, Iida and Ishihara studied the effect of light induced force microscopy of resonant quantum dot systems [64–66]. Some experiments have also been carried out, to our knowledge the first being by Zhu, et al. in 1997, which examined the effect of the shear force feedback signal as a function of tip-sample separation both when the laser light was coupled into the probe and when the probe was illuminated from the side [67]. The following year, the same group published a follow up paper in which they reported observing a somewhat higher optical force induced effect on an Al coated tip than on a bare SiO₂ tip and a significantly higher force when using a high dielectric constant substrate as opposed to a low dielectric constant substrate. These authors did not notice any effect when a metal substrate was used [68].

More recently, Satoh, et al. detected the variability of the light transmission through a checkerboard structure of Cr patches on a glass substrate using optical force induced changes in the resonance frequency and dissipation of the probe [23]. Additionally, Kohoutek, et al.

performed measurements claiming to measure the optically induced force in a bowtie antenna using an AFM probe [69].

These studies clearly demonstrate the possibility to exploit optically induced forces for typically NSOM operating conditions. However what is lacking is a systematic study confirming the origin of these forces and thus the promise they hold to complement or replace standard NSOM measurements. Such a study is the focus of this dissertation.

3.3 Other indirect (thermal) effects on the cantilever

Of particular interest to this report are thermally induced effects on the probe and in particular thermally induced tip elongation. The feedback for SPM systems is intended to keep the tip-sample separation constant during a scan. If a probe elongates, then the feedback uses a piezo to retract the probe (or sample) in order to maintain that constant separation; the topography is actually determined by changes in the properties of this piezo. Thus, if the degree of probe elongation varies spatially across the sample, this will introduce spurious topographic features into the measurement, similar to the topography of light discussed in Chapters 4 and 5.

For example, in illumination mode, light is coupled into the probe and excites the sample. In order to achieve high spatial resolution a sharply tapered metal coated probe is used. Because the small throughput in the aperture (typically on the order of $10^{-5} - 10^{-6}$), almost all of the incident radiation is either reflected back or absorbed in the metal coating. This can cause a variety of thermally induced effects, such as: tip elongation which will tend to reduce light throughput, aperture expansion which will increase light throughput, changes in the metal coating reflectivity, and of course, for sufficiently high powers, even optically-induced damage can occur [70,71]. Indeed, a number of measurements have been performed and models proposed

to determine the temperature increase in the tip as light is coupled into it [27,72]. Notably, a collection mode experiment similar to the one in our studies was undertaken by Lienau, et al. [25] and, as such, will be discussed in more detail in section 5.1.

Another related study was conducted more recently, which examined the effect of tip heating over a laser diode face on the topography and lateral friction, which the authors credited to thermally induced bi-material effects [73]. The effect of illuminating the probe from the side was also studied and the observed optically induced effects were credited optical binding [68]. Clearly, the question arises of as to the true origin or origins of the observed optically induced effects in topography measurements.

3.4 Other Optically Induced Effects

Thus far in this chapter we have discussed scanning probe techniques that may be affected by optically induced forces, either directly or indirectly. Here we briefly outline some tangentially related ideas.

In each of the cases discussed, a probe is used to locally sample a physical property but the true detection is done via the feedback based on the effect of different force gradients on the tuning fork or cantilever on which the probe is attached. The only purpose of the probe is to sample the property of interest on a finer scale and perhaps with more accessibility than might otherwise be possible. However, the tuning fork or cantilever is the critical detection component. Thus, in certain scenarios one can bypass the probe and just use the tuning fork or cantilever itself as a detector of electromagnetic radiation., which is of particular interest currently for the detection of THz radiation [74,75].

In addition to optically induced effects present on the probe itself, the laser bouncing off the back of the cantilever can also affect the cantilever deflection [76,77]. It was shown that even for an excitation power of $100\ \mu W$, the uncoated probe studied deflected by $30\ nm$ when excited from one side whereas the metal coated probe deflected over $3\ \mu m$; standard commercially available probes were used in both cases [76]. These studies showed that the optical effects on uncoated SiN cantilevers are dominated by radiation pressure whereas for metal coated SiN structures the major influence is thermal, the cantilever deflecting primarily due to bi-material effects. These observations provided an interesting opportunity to optically modulate the cantilever with light [76], to optically increase [78] or decrease [79] the cantilever quality factor or even change the cantilever spring constant [80], which can, for example, improve the time response without sacrificing the force sensitivity [79] or increase the force sensitivity [80].

A related measurement technique is the so-called photonic force microscopy [81–84]. In this technique, the fluctuations of the particle position within a three-dimensional standard Gaussian trap are monitored using a quadrant photodetector. By measuring the statistics of these deflections, the potential well can be calibrated and then used to measure additional forces acting on the particles. By scanning the particle relative to the sample, the forces may be measured in a manner analogous to atomic force microscopy [82,84,85]. Near field measurements may also be performed, for example by exciting the sample with a second laser with a different frequency and significantly lower power in total internal reflection and then measuring the light scattered off the particle in the far field [85]. This microscopy is different from the optically induced forces we discuss in this dissertation because it relies on Brownian motion to measure the statistics of

fluctuations in particle locations in order to determine the external force acting on the test particle.

Everything we have discussed in this chapter has dealt with the detection or unintended influence of light which manifests itself via changes in the tuning fork or cantilever resonant conditions. For specific material systems, the interaction of light with a sample may induce permanent changes to the sample topography, such as the local formation of the oxide of the sample material. By performing standard AFM scans before and after irradiation of light, it is possible to indirectly measure the light intensity in locations where it is intense enough to cause said oxide formation simply by looking at how the topography was affected by the light irradiation [86].

Last and perhaps most importantly, extensive theoretical and experimental work has been undertaken in the field of optical forces acting on small particles, particularly evanescent optical fields excited in a TIR illumination condition acting on spherical particles [87–90]. We emphasize this last example because often an NSOM probe is modeled as a small sphere and illumination in TIR is common for this microscopy. Such calculations, which appeared in the early 1990's, show that reasonable illumination intensities should produce forces that are measureable by an atomic force microscopy probe [61,62]. Thus, it is interesting to understand the reasons for this dearth of experimental evidence. Suffice to say, the presence of optically induced forces has been searched for in the past but many questions remain unanswered. The scope of this dissertation is to clarify some of these unexplored possibilities, as described below.

The first striking omission of earlier studies is a careful examination of optically induced forces in a purely transparent dielectric system. Such system has the advantage of eliminating, or

at least greatly reducing, any thermal effects. Indeed, our preliminary measurements described in Sec. 5.1 were done on a pure dielectric system for exactly this reason.

Second, we have not found a discussion of the effects these forces have in the practice of NSOM measurements. Indeed, these forces are typically implicitly or explicitly assumed to not be present. However, as we show in Chapter 4, such effects may be readily observed for even moderate illumination intensities in transparent dielectric systems.

Third, in Chapter 5, we demonstrate the quantitative determination of the spatially resolved optically induced force acting on a scanning probe. A by-product of this process is the possibility to determine the tip-sample separation, which we find as a consequence of controllably varying the input light intensity. This simple technique allows us to bypass the need for complex external measurement schemes [8,9].

Fourth, the applications of these optically-induced forces have, thus far, been limited. Even for the relatively well studied Kelvin probe force microscopy, their description has been limited to the realm of curiosity rather than practical utility. In this dissertation, we address this utility. In addition, we demonstrate in Chapter 6 that standard NSOM and optical force based measurements are sensitive to different components of the complex 3D field. Thus, by measuring simultaneously these two different interactions of the EM field with the probe, more information about that field distribution is obtained without increasing the complexity of the experiment.

In Chapters 4-6, we address the measurement of optically induced force gradient effects via the shift in the probe resonance frequency. In Chapter 7 we explore the possibility to drive optically the probe oscillation, which allows measuring the optically induced force in addition to its local gradient. Finally, we note that while different groups attribute their observed optically

induced effects to various physical phenomena, the question still remains as to what exactly a scanning probe is measuring: thermally induced effects, optical gradient effects or optical binding effects. This issue is addressed in Ch. 7.

4 ARTIFACTS DUE TO OPTICALLY INDUCED FORCES (OIF)

Having completed a review of previously reported optically induced forces in scanning probe microscopy mostly related to Kelvin Probe Force Microscopy, we turn our attention to the presence and influence of unintended optical force effects in AFM and NSOM.

4.1 Forces due to Electromagnetic Fields

The focus of this dissertation is direct optical field induced effects on scanning probes. The force due to an arbitrary incident electromagnetic field on an arbitrary object may be found starting with the first two Maxwell's equations, which in Gaussian units for an object in vacuum are given by:

$$\begin{aligned}\nabla \times \vec{E} &= -\frac{1}{c} \frac{\partial \vec{H}}{\partial t} \\ \nabla \times \vec{H} &= \frac{1}{c} \frac{\partial \vec{E}}{\partial t} + \frac{4\pi}{c} \vec{j}\end{aligned}\tag{4-1}$$

where $\vec{E}$ is the electric field, $\vec{H}$ is the magnetic field, c is the speed of light, and $\vec{j}$ is the total current density. Operating on Maxwell's first equation with $\times \vec{E}$ and on Maxwell's second equation with $\times \vec{H}$, one obtains after some manipulation [91]

$$\nabla \cdot \frac{1}{4\pi} \left[\vec{E}\vec{E} - \vec{H}\vec{H} - \frac{1}{2} \left(|\vec{E}|^2 + |\vec{H}|^2 \right) \vec{I} \right] = \frac{d}{dt} \frac{1}{4\pi c} \left[\vec{E} \times \vec{H} \right] + \rho \vec{E} + \frac{1}{c} \vec{j} \times \vec{B} \tag{4-2}$$

where ρ is the total charge density, η is the free space impedance, $\vec{I}$ is the unit tensor, $\vec{B}$ is the magnetic field density and $\vec{E}\vec{E}$ denotes the outer product of the electric field. The expression square brackets in the left hand side of the Eq. (4-2) is the so-called Maxwell's stress tensor, $\vec{T}$, which reads

$$\vec{T} = \frac{1}{4\pi} \left(\vec{E}\vec{E} - \vec{H}\vec{H} - \frac{1}{2}(|E|^2 + |H|^2)\vec{I} \right). \quad (4-3)$$

Integrating Eq. (4-2) over an arbitrary volume, V , containing both the sources ρ and $\vec{j}$, gives

$$\int_V dV (\nabla \cdot \vec{T}) = \int_V dV \left(\frac{d}{dt} \frac{1}{4\pi c} [\vec{E} \times \vec{H}] \right) + \int_V dV \left(\rho \vec{E} + \frac{1}{c} \vec{j} \times \vec{B} \right) \quad (4-4)$$

The last term represents the force law for a distribution of charges and currents satisfying the charge conservation law

$$\nabla \cdot \vec{j} + \frac{\partial \rho}{\partial t} = 0. \quad (4-5)$$

Using Gauss' theorem, one can replace the stress tensor volume integral with an integral over the bounding surface of the volume. In this case

$$\int_V dV (\nabla \cdot \vec{T}) = \int_S da (\vec{T} \cdot \hat{s}), \quad (4-6)$$

where $\hat{s}$ is the unit vector perpendicular to the surface, S , which encloses the volume, V .

With this, we may now write

$$\int_S da (\vec{T} \cdot \hat{s}) = \frac{d}{dt} [G_{field} + G_{mech}], \quad (4-7)$$

which represents the conservation law for linear momentum. Here G_{field} and G_{mech} are respectively the field and mechanical momentum. Integrating Eq. (4-7) over one oscillation period gives

$$\langle F \rangle = \int_S da (\langle \vec{T} \rangle \cdot \hat{s}) \quad (4-8)$$

since the field momentum averages to zero and $F = dG_{mech}/dt$. Here $\langle \rangle$ denotes the time average.

Equation (4-8) is a general result which may be used to calculate the mechanical force acting on an arbitrary object surrounded by an arbitrary surface illuminated by an arbitrary electromagnetic field. We note that the electromagnetic field entering in the force evaluation in Eq. (4-8) is calculated self-consistently; it represents the superposition of the incident and scattered fields. Note that this derivation also assumes that the object does not deform under the action of the electromagnetic field. Otherwise electro-restrictive and magneto-restrictive forces must be included; likewise, Eq. (4-6) will not be valid [91].

From Eq. (4-8) we see that the force on an object induced by an electromagnetic field is entirely determined by the fields at the surface of a volume enclosing the object with no dependence on the medium's properties. This surface may be located on the surface of the object itself or far away from it. Of course, the material properties do play a role, as they determine self-consistently the electromagnetic fields used to calculate the force.

In the typical NSOM experiment, a sharp probe is used to sample the local field distribution. Due to its small size, the dipolar approximation, which is valid for particles much smaller than λ/n where n is the refractive index of the embedding medium, may be used. When such a particle is exposed to a time harmonic field, Eq. (4-8) simplifies to [92]

$$\langle F_i \rangle = \frac{1}{2} \Re \left[\alpha E_j \frac{\partial E_j^*}{\partial x_i} \right] \quad (4-9)$$

where $\Re$ denotes the real part, α is the particle polarizability, $\langle F_i \rangle$ is the time averaged force of the i^{th} Cartesian component and E_j are the complex-amplitude electric field components. Summation over repeated indices is implied. The particle polarizability is then given by

$$\alpha = \alpha_0 \left[1 + \frac{2}{3} i k^3 \alpha_0^* \right] / D \quad (4-10)$$

where * stands for the complex conjugate and $\alpha_0 = a^3 (\epsilon_p - \epsilon_s) / (\epsilon_p + 2\epsilon_s)$ where a is the radius of the dipole, and the dielectric constants are defined in Figure 4-1. The correction factor D is given by $D=1$ for dielectric particles and $D = 1 + 4/3 (k^3 \Im(\alpha_0)) + 4/9 (k^6 |\alpha_0|^2)$ for metallic particles where $\Im$ stands for the imaginary part [93,94].

Figure 4-1: Schematic of dipole over a surface.

Assuming plane wave illumination is incident on the dipole, the electric field can be written as

$$\vec{E} = \vec{E}_0 e^{i\vec{k} \cdot \vec{r}} \quad (4-11)$$

where k is the wavevector in the direction of propagation. This plane wave may be either propagating or evanescent. Substituting Eq. (4-12) into Eq. (4-10), we obtain

$$\langle F \rangle = \frac{1}{4} \Re(\alpha) \nabla |\vec{E}_0|^2 + \frac{1}{2} \vec{k} \Im(\alpha) |\vec{E}_0|^2 - \frac{1}{2} \Im(\alpha) \Im(\vec{E}_0 \cdot \nabla \vec{E}_0^*). \quad (4-12)$$

We note the time averaged force acting on the dipolar particle can be decomposed in three different contributions, which, from left to right, are the gradient force, the radiation pressure

force and a possible spin force. For our discussion in the following it is important to note that the real part of the polarizability contributes to the gradient force whereas the imaginary part contributes to the scattering-plus-absorption force.

The incident electric field in Eq. (4-12) may be decomposed into

$$\vec{E} = (\vec{E}_{0x}, 0, \vec{E}_{0z}) e^{i\vec{k} \cdot \vec{r}} \quad (4-13)$$

for p-polarized light and

$$\vec{E} = (0, \vec{E}_{0y}, 0) e^{i\vec{k} \cdot \vec{r}} \quad (4-14)$$

for s-polarized light. In this case, the force exerted on the dipolar particle by the electromagnetic field becomes [93]

$$\begin{aligned} F_x &= \frac{1}{2} \Re \left[\alpha (ik_x)^* \left(|E_{0x}|^2 + |E_{0z}|^2 \right) \right] \\ F_z &= \frac{1}{2} \Re \left[\alpha (ik_z)^* \left(|E_{0x}|^2 + |E_{0z}|^2 \right) \right] \end{aligned} \quad (4-15)$$

for p-polarized light and

$$\begin{aligned} F_x &= \frac{1}{2} \Re \left[\alpha (ik_x)^* \left(|E_{0y}|^2 \right) \right] \\ F_z &= \frac{1}{2} \Re \left[\alpha (ik_z)^* \left(|E_{0y}|^2 \right) \right] \end{aligned} \quad (4-16)$$

for s-polarized light. The above equations are valid for particles embedded in an external medium with a uniform refractive index, i.e. when the particle is far from any surface, such that multiple scattering effects do not need to be taken into account. These equations may be used for either propagating or evanescent incident plane waves.

When performing NSOM experiments, the probe will typically be close to the sample's surface in order to interact with the evanescent fields. In this case, we must account for multiple

scattering between the dipole (tip) and the sample surface. This interaction introduces a correction to the force due to optical binding which occurs between the probe and surface. In the case of a purely dielectric sample, the force of the electromagnetic field on the dipole is given by [94]

$$\begin{aligned}
F_x &= \frac{1}{2} \Re \left[4\alpha z_0^3 (ik_x)^* \left(\frac{2|E_{0x}|^2}{8z_0^3 + \alpha\Delta} + \frac{|E_{0z}|^2}{4z_0^3 + \alpha\Delta} \right) \right] \\
F_z &= \frac{1}{2} |E_{0x}|^2 \Re \left[\left(\frac{8\alpha z_0^3 (ik_z)^*}{8z_0^3 + \alpha\Delta} + \frac{12|\alpha|^2 z_0^2 \Delta}{|8z_0^3 + \alpha\Delta|^2} \right) \right] + \\
&\quad + \frac{1}{2} |E_{0z}|^2 \Re \left[\left(\frac{4\alpha z_0^3 (ik_z)^*}{4z_0^3 + \alpha\Delta} + \frac{6|\alpha|^2 z_0^2 \Delta}{|4z_0^3 + \alpha\Delta|^2} \right) \right]
\end{aligned} \tag{4-17}$$

for p-polarization and

$$\begin{aligned}
F_x &= \frac{1}{2} |E_{0y}|^2 \Re \left[\frac{8\alpha z_0^3 (ik_x)^*}{8z_0^3 + \alpha\Delta} \right] \\
F_z &= \frac{1}{2} |E_{0y}|^2 \Re \left[\left(\frac{8\alpha z_0^3 (ik_z)^*}{8z_0^3 + \alpha\Delta} + \frac{12|\alpha|^2 z_0^2 \Delta}{|8z_0^3 + \alpha\Delta|^2} \right) \right]
\end{aligned} \tag{4-18}$$

for s-polarization. Here $\Delta = (\varepsilon_s - \varepsilon_d) / (\varepsilon_s + \varepsilon_d)$, which is the Fresnel reflection coefficient at the surface. Because the interface is assumed to be a lossless dielectric, Δ is a real quantity.

To this point we have discussed how to calculate the force on arbitrary particles as well as of a dipole either embedded in a homogeneous medium or in the presence of a dielectric interface. Naturally, the dipole approximation is only valid when the fields across the dipole can be considered constant, a constraint which, as noted earlier, is met, for example, when a plane wave illuminates a particle which is much smaller than the wavelength. For larger particle or for

more complex local field distributions, as are commonly found in typical NSOM experiments, it becomes necessary to take higher order moments into account to accurately compute the force [93,95,96]. Likewise if the dipole has a finite magnetic dipole moment, terms accounting for both the magnetic dipole moment as well as the coupling between the electric and magnetic dipole moments must additionally be computed [88,95]. In many cases, such analytical formulations become intractable and one has to turn to numerical modeling approaches.

4.2 Optically induced forces (OIF)

Now that we have given a theoretical description of direct optically induced forces, we turn our attention to experimental observations of such optically induced forces. Since their invention, both aperture- and scattering-based near field scanning optical microscopes (a-NSOM and s-NSOM) have been used as optical imaging tools capable of sub-diffraction limited resolution [37,41]. Generally speaking, such resolution is realized by either exciting or collecting radiation from a very small region of a sample. To do so, a physical probe is brought in the proximity of the sample and used to couple near-field onto far-field radiation that can then be detected remotely. However, extraction of a pure near field signal is complicated by the presence of a background signal, which, in most modalities of near field imaging, can introduce artifacts [39,97]. Techniques to correct or eliminate these artifacts have been extensively studied [35,37,41]. We discussed this in detail in section 2.4.

Like other scanning probe microscopies, NSOM relies on the interaction of a probe with a sample to perform its measurement. Intrinsic to these techniques is the introduction of undesired interactions, which are often referred to as imaging artifacts as they complicate or completely obscure the measurement of the desired signal. Perhaps the most well-known are the

z-motion artifacts discussed in section 2.4.1. In this chapter we discuss an additional imaging artifact due to optically induced forces that may be present in the acquisition of collection/scattering mode NSOM images. Although we discuss optically induced forces present in typical a-NSOM scans, similar artifacts should also be considered in s-NSOM where a sharp metal probe is used in order to follow the sample profile and to enhance the local field contribution to scattering.

We illustrate the effect of optical force-induced artifacts in a few typical NSOM measurements. For this, two different samples were mounted on a triangular prism and illuminated in total internal reflection (TIR) by a laser with a 532 nm wavelength. An uncoated tapered optical fiber probe of $\sim 20\text{ nm}$ in diameter is scanned with feedback engaged to collect topographic maps in the equivalent of collection mode NSOM. A dielectric probe was chosen in order to eliminate other possible effects such as thermally induced melting/damage of the probe [98] or tip elongation [25].

The subtle influence of OIF in topographic images is illustrated in Figure 4-2. In this example the sample consists of a thin Au film evaporated onto a quartz substrate. The film has been annealed to open up pits in the coating. Three measurements were performed across the edge of the Au film: first one without illumination to determine the “real” surface topography, z_{off1} , a second one, z_{on} , with the surface is illuminated in TIR mode with a peak incident intensity of $\sim 0.25\text{ mW}/\mu\text{m}^2$, and finally a third topography z_{off2} with the illumination turned off again. The “real” topography, z_{off1} , (dash-dotted line in Figure 4-2) reveals the transition between the quartz surface and the Au film which occurs at $\sim 0.6\text{ }\mu\text{m}$. The solid line in Figure

4-2 is the computed difference $\delta Z_1 = Z_{on} - Z_{off1}$, which provides a measure of the optically induced topography. To gauge the scan repeatability, noise and any sample damage, we have also evaluated $\delta Z_2 = Z_{off2} - Z_{off1}$, the difference between the two “real” topography scans (dashed line). This difference is clearly within the noise indicating good scan repeatability and the absence of significant optically induced transformations in the sample. However, at the edge of the Au film, the tip retracts $\sim 20 \text{ nm}$ from where we expect. Note that often the locations of interest are such regions of high local field intensity which occur at discontinuities in sub-wavelength metallic structures, an edge in our example. As one can see in Figure 4-2, these large field enhancements can affect even dielectric probes illuminated at low intensity.

Figure 4-2 “Real” surface topography (dash-dotted line) of a Au film deposited on quartz which has been annealed to open up pits in the coating. The solid line is the difference between topographies recorded with and without illumination. The dashed line is the difference between two scans of the real topography performed before and after illumination. Due to optical forces, the tip is pushed ~ 20 nm away at the edge of the Au film.

Physically, the tip retraction occurs because the feedback works to maintain a constant phase shift on the tuning fork excitation signal and, thus, a constant force gradient on the probe. In our case, this is equivalent to saying that the feedback maintains a constant resonance frequency shift on the tip [99]. In the presence of the attractive, optically-induced force, this causes the tip to retract in order to reduce the contribution of Casimir (van der Waals) forces on the probe, which are also attractive. In terms of induced frequency shifts, this means $\Delta f_{C_off} = \Delta f_{C_on} + \Delta f_{opt}$, where Δf_{C_off} and Δf_{C_on} represent the frequency shifts due to the

Casimir forces when the incident radiation is turned off and on, respectively and Δf_{opt} is the frequency shift due to OIF [99]. For the relatively large oscillation amplitudes ($\sim 20 \text{ nm}$) in our measurements, the frequency shift relates to the force F acting on a probe as $\Delta f = -(f_0^2 / kA^2) \int_0^{1/f_0} AF(x_{tip}) \cos(2\pi f_0 t) dt$ where f_0 is the resonance frequency, k is the spring constant, A is half of the total tip oscillation amplitude, t is time and $x_{tip} = [x_{CL_b}, y_{CL_b}, z_{CL_b} + A \cos(2\pi f_0 t)]$ where $(x_{CL_b}, y_{CL_b}, z_{CL_b})$ is the coordinate at the base of the cantilever [100].

We emphasize that the optically induced effect observed in Figure 4-2 is confined at the metal edge. Due to this localization, it is difficult to identify its presence by simply examining the topography data. The influence of optical forces, however, can be clearly isolated when analyzing relative measurements performed with and without illumination, as we have done here.

In the example discussed, metal is still present and, therefore, one cannot completely eliminate the possibility of thermally induced effects such as thermal expansion in the sample. To emphasize the sensitivity of NSOM scans to OIF and confirm that such topographical artifacts have an electromagnetic rather than thermal origin, we present another example in which we have completely removed metal from the experiment. In this case, a nanostructured dielectric GaP sample was scanned with the same uncoated dielectric AFM probe. We again performed three successive measurements: first of the “real” topography, z_{off1} , then with the sample illuminated in TIR mode, z_{on} , and finally we repeated the scan of the “real” topography, z_{off2} . The three-dimensional plots in Figure 4-3 show the “real” topography, z_{off1} , while the color maps represent the optically induced topographies, $\delta z_1 = z_{on} - z_{off1}$, in Figure 4-3(a) and

the control, $\delta z_2 = z_{off2} - z_{off1}$, in Figure 4-3(b). Again, one can observe large increases in the tip sample separation ($\sim 50 \text{ nm}$) along the sharp edges of the sample even with a peak incident illumination intensity of only $\sim 2 \text{ mW}/\mu\text{m}^2$. The reduced color variations in Figure 4-3(b) clearly demonstrate that the effect visible in Figure 4-3(a) is not due to drift between scans or other measurement variability.

Figure 4-3 The effect of optically induced forces on a nanostructured GaP sample measured with an uncoated NSOM tip. (a) Optically induced topographies, $\delta z_1 = z_{on} - z_{off1}$ and (b) control difference $\delta z_2 = z_{off2} - z_{off1}$ overlaid on the "real" topography, z_{off1} . Note that the largest artifacts induced by optical forces are located at the edges where the strength of the local z directed intensity gradient is the highest.

4.3 Discussion of the presence of OIF in different modalities

Having demonstrated the existence of these optical force effects, we comment on how they can be detected and perhaps eliminated from other measurements. The experiments in this chapter illustrate the effect of optical forces in collection mode NSOM for a tuning fork based probe operating in normal force intermittent contact mode using phase feedback. Similar effects should also be present in amplitude feedback, since the optically induced forces act to shift the resonance frequency which will change both the tip resonance frequency amplitude and phase. It has been shown that the influence of optical forces is detectable also in a cantilever based NSOM operating in non-contact mode by observing the cantilever deflection [69] as well as in intermittent contact mode using either frequency shift or dissipation feedback mechanisms [23]. The effect of optical forces on scanning probes is always present but, given the variety of available probes, operating modalities, and feedback mechanisms, it is conceivable that the strength of these effects may depend on the specific conditions of operation. For instance, one can expect the influence of optical forces to be diminished in contact mode NSOM due to the much stronger surface forces acting on the probe.

For tuning fork based feedback we expect that the influence of optical forces could be reasonably eliminated by performing constant height mode (CHM) scans where feedback is turned off; however, a cantilever based tip may still deflect under the action of these forces, even when the feedback is turned off. Depending on the exact experimental conditions, OIF may have noticeable effects even hundreds of nanometers or more from the surface. Of course, OIF becomes negligible when reducing the illumination intensity (in our case the detection threshold is on the order of piconewtons [99]). In NSOM practice however, large local intensities are

usually desired in order to have a detectable signal through a small aperture or scattered from a sharp tip. Thus, avoiding the influence of optical forces acting on a scanning probe in the presence of external illumination may not be possible and all that one can do is to quantify the effect of OIF. In constant-gap mode (CGM) where feedback used to regulate the tip-sample separation, this may be done by analyzing the topography maps [99]. In CHM, one can read the signal normally used to regulate the tip-sample separation and compute the force and thus infer the tip retraction (deflection) or shift in the resonance frequency. If this resonance frequency shift is large enough, the oscillation amplitude of the probe will also be affected (in general reduced) because the tuning fork compliance at the driving frequency will change. The optical signal must then be corrected, which may require the use of approach curves at locations of interest. Because of the large field gradients, similar effects should be expected in aperture NSOM operated in illumination mode. However, for metal coated tips, this modality has the additional complication of thermally-induced tip elongation [25].

Interestingly and arguably most notably, the effect of these forces may be significant but not readily apparent in the topographical scans of structured samples because the differences in these scans are rather localized and can be easily obscured by somewhat larger “real” topographic variations. However, substantial effects can be observed by subtracting the “real” topography collected without illumination. Because of these modifications in the effective topography followed by the scanning probe, the optical image (the NSOM data) is collected not at the intended tip sample separation, but rather from a different location in the strongly spatially varying 3D field distribution surrounding nanostructures.

In conclusion, we have shown that the optically induced forces acting on an NSOM probe are strong enough to modify the tip-sample separation in regions of sufficiently strong field intensity. Failure to take this effect into account results in a significant underestimate of the peak field enhancement as well as in recording erroneous topographic maps. These findings are particularly important for the practice of s-NSOM, which relies on following closely the sample topography with a sharp metal tip in order to generate images of the optical near fields. As pointed out in Ref. [37] several years ago, “*for near field microscopy to become a reliable imaging tool, it is indispensable that inherent artifacts should be carefully accounted for*”. We have shown here that, in addition to eliminating artifacts related to background signals, a correct interpretation of scanning probe microscopy measurements also requires understanding the influence of optical forces.

5 FORCES IN NEAR FIELD SCANNING OPTICAL MICROSCOPY (NSOM)

Having demonstrated and qualified the presence of optically induced forces in AFM and NSOM in Chapter 4, we now address the problem of the quantitative measurement of their strength. In section 2.6 we discussed a subset of the forces present in SPM scans. In this chapter we show how knowledge of those forces allows us to characterize the optically induced forces acting on the probe. As a consequence of the analysis, we will also demonstrate that using this simple experimental setup, we can determine the tip-sample separation, something which otherwise requires comparatively complex setups to determine [8,9].

5.1 Optical forces in propagating fields

To measure the influence of the optical forces on a scanning probe, we performed scans over the core of a single mode fiber with its exit surface cleaved and polished normal to the optical axis. Laser radiation of several different powers was coupled into an optical fiber with an effective mode diameter of the fiber of $3.5\ \mu m$. In this experiment the probe was an uncoated tapered glass capillary tube with a $100\ nm$ diameter aperture mounted on a Nanonics MV4000 scanner. Because optically induced forces affect the scanner's feedback mechanism, topography scans are sensitive to optical radiation impinging on the tip. By separately measuring the "real" surface topography, we extract the optically induced contribution to the measured topography, as illustrated in Figure 5-1.

Figure 5-1 (Color online) Perceived topography over the core of a single mode optical fiber when (a) 24 and (b) 16 mW of 532 nm laser light is coupled into fiber. The inset in (a) is the side view of the probe; the scale bar corresponds to 100 μm . The inset in (b) shows the measured topography of the fiber face. The blue dashed circles indicate the location of the fiber core which is $\sim 2.2 \mu m$ in diameter. The arrow indicates the orientation of the tip during scanning.

Several features of these optical topographies are worth noting. First, we see that the effect of optically induced forces manifests itself as an increase of the tip sample separation. This is because the light impinging on the probe generates a negative shift in the tuning fork resonance frequency whose strength depends on the 3D field distribution and its interaction with the probe. The feedback compensates for the additional optically induced frequency shift by retracting the tip to reduce the contribution of surface forces, thereby maintaining a constant resonance frequency shift on the tuning fork. Thus, we observed an increase in the measured height or, in other words, a perceived “topography of light”. Additionally, we notice that the effect of the light on the probe is strongest in the upper left corner of the scan, which corresponds to tip positions past the core of the fiber. This somewhat counterintuitive observation can be explained by realizing that the optical interaction with a beam emerging from the fiber

propagates over an extended volume and its strength depends on the size, shape, and orientation of the probe. As seen in the inset of Figure 5-1(a), the tip has a conical shape and is also tilted with respect to the scanning plane across the interface. Therefore, a larger volume of the probe is exposed to light when the tip has already passed over the core, which results in a greater optically induced force. The dependence on the volume of interaction is an important characteristic of the mechanical action of light and will be analyzed in detail later.

As was mentioned in section 3.3, this experiment is similar to the one presented by Leinau, et al [25]. In both cases an SPM probe was scanned over an illuminated spot, in our case the face of the single mode optical fiber and in their case the end facet of a laser, and the topography signal recorded. They scanned the sample with three probes: a fully aluminum coated tip, a standard tip and an uncoated tip for output powers up to 3 mW , after which point the fully aluminized tip is damaged. They find a significant retraction of the feedback piezo that, for the metal coated probes, depended linearly on of the incident power. However, a negligible effect was noted in the case of an uncoated probe. The observed retraction was attributed to thermally induced tip elongation [25]. One significant difference between the experiment described here and the one in Ref. 25 is that their SPM operated on shear-force feedback whereas ours was based on normal-force feedback [61]. Because a shear-force feedback system spends most of its oscillation cycle much closer to the sample than a normal-force feedback system and because the detection of optical gradient forces requires those forces to be on the order of the competing van der Waals/Casimir forces which decay rapidly from the surface, it is entirely possible that our measurement approach is more sensitive to the gradient of the optically induced force. In support of this observation we also note that in Ref [25] the effect of optical forces was

not observed on the dielectric probe even for significantly higher illumination intensities than the ones used in our experiments.

5.2 Oscillating probe and frequency detuning

To model the effect illustrated in Figure 5-1, we may interpret the behavior of our tip as a damped driven harmonic oscillator [15]. In the proximity of an interface, the probe is in general affected by different types of interaction forces that cause the resonance frequency of the tuning fork to shift and introduces an additional damping mechanism [15]. These two effects result in changes of both the amplitude and the phase of the tip oscillation relative to the driving signal; the system's feedback acts to maintain this phase constant.

In our experiment, the only important external contribution to the probe damping is due to the energy dissipation as the probe contacts the surface during its oscillation cycle. The scans were operated such that little energy is lost due to contact with the sample surface when the light is off, and this is further reduced when the light pushes the tip away from the surface. Consequently, we can neglect damping and analyze the probe oscillation based on the induced resonance frequency shift $\Delta\omega$ in a harmonic oscillator of mass m that has a resonance frequency $\omega_0^2 = k / m$ determined by its spring constant k . The dependence between the frequency shift $\Delta\omega$ and the force acting on the tip $F_{tip}(z)$ can be calculated as a first order perturbation using the Hamilton-Jacobi approach [18,100]; in this case $\Delta\omega = -(\omega_0 / kA^2) \langle F_{tip}(z)x(t) \rangle$ where $x(t)$ is the unperturbed motion of tip in the absence of $F_{tip}(z)$, and averaging is performed over an entire oscillation cycle. If the force gradient $k_{tip} = -\partial F_{tip} / \partial z$ is constant during the oscillation cycle, the frequency shift $\Delta\omega = \omega_0 k_{tip} / (2k)$

is simply proportional to the force gradient. However, because our scanning probe operates in intermittent contact, the amplitude of oscillation A can be of the order of tens of nanometers. Consequently, k_{tip} can vary by orders of magnitude during one oscillation and a more precise evaluation of the frequency shift is necessary, as detailed in the following.

5.3 Forces acting on probe

In the experiment described above, the dominant interaction between the surface and the probe is through dispersion forces [47]. When the tip is also under the influence of an external electromagnetic field, additional forces due to radiation pressure, gradient of intensity or phase, optical binding, photophoretic forces (local heating) and photoinduced stress may be present [23,68]. To isolate the optical effect, for this particular experiment we used a purely dielectric system such that significant thermal, electrostatic and magnetic effects can be avoided. Thus, in our experiments, the two main external contributions to the total force acting on the oscillating probe are the retarded van der Waals force F_W and the mechanical action of light F_{opt} .

When the feedback is engaged, a constant resonance frequency shift

$$\Delta\omega_{opt} = \Delta\omega_W - \Delta\omega_{W_{opt}} \quad (5.1)$$

is maintained. In Eq. (1), $\Delta\omega_W$ and $\Delta\omega_{W_{opt}}$ represent the frequency shifts on which the feedback operates when the incident radiation is turned off and on, respectively. To derive analytic equations for these terms, we model the tip-sample geometry as a silica sphere in air interacting with a silica plane. This approach is common for estimation of van der Waals forces in AFM and is valid for probes with paraboloidal tips. In our case this approach is valid because

the radius of tip is quite large ($\sim 150 \text{ nm}$) and, therefore, the influence on the bulk of the tip is negligible at typical tip-sample separations. We emphasize that this model is only used for the estimate of the van der Waals forces. Accounting for the large variation of the interaction forces over the course of one tip oscillation cycle, the resulting shift in the resonance frequency is obtained as [100]

$$\Delta\omega_W = \frac{\omega_0}{kA} \frac{2\pi RB}{3z^3} \left[F_1^{3,0.5} \left(\frac{-2A}{z} \right) - F_2^{3,1.5} \left(\frac{-2A}{z} \right) \right] \quad (5.2)$$

where $F_a^{b,c}(z)$ is the hypergeometric function, B is the retarded Hamaker constant [46], R is the radius of the sphere and z is the closest tip sample separation. The expression for $\Delta\omega_{W_opt}$ is the same as Eq. (2) except that z is now replaced with $z + \delta z$ where δz describes the optically induced topography shown in Figure 5-1. The retarded van der Waals force is estimated to be of the order of 10^{-8} N near the surface and down to 10^{-14} N at one micron away from the surface.

As the distribution of the electromagnetic field emerging from the fiber changes only slightly over a distance of a few microns, the longitudinal gradient of the optical force is nearly constant over the range of tip oscillations considered. This assumption will be confirmed later by our numerical calculations. Thus, $\Delta\omega_{opt}$ is found to be

$$\Delta\omega_{opt} = -\frac{\omega_0}{2k} \frac{\partial F_{opt}}{\partial z} = \frac{-\omega_0}{2k} \alpha I \quad (5.3)$$

where α is a constant that measures the force exerted on the tip by a unit of optical power coupled out of the fiber and I is the local field intensity at the point (x, y) .

5.4 Optical force distribution

Using Eqs. (5.1)-(5.3), we obtain a direct relationship between the optically induced forces and the resulting changes in the dispersion force acting on the tip. Alternatively, we can determine the strength of the optically induced force gradients if the strength of the van der Waals contributions are known before and during the light irradiation, i.e. for separation distances z and $z + \delta z$:

$$\frac{\partial F_{opt}}{\partial z} = \frac{-4\pi RB}{3A} \left[\frac{1}{z^3} F_W^3 - \frac{1}{(z + \delta z)^3} F_{W_{opt}}^3 \right] \quad (5.4)$$

where

$$F_W^n = F_1^{n,0.5}(-2A/z) - F_2^{n,1.5}(-2A/z) \quad \text{and}$$

$F_{W_{opt}}^n = F_1^{n,0.5}(-2A/(z + \delta z)) - F_2^{n,1.5}(-2A/(z + \delta z))$. Because the gradient of the dispersion force is a nonlinear function of the tip sample separation, we require knowledge of the initial tip sample separation at closest approach in order to determine the gradient of the optical force. Obtaining this quantity, which is typically not known, usually requires external measurements or *a priori* assumptions [8,9,16]. Here we exploit the fact that the gradient of the optical force is linear with respect to the field intensity $I(x, y)$ at each point during the scan. Thus, one can use Eq. (5. 4) to compute the initial tip sample separation z at each location in the scan by evaluating the ratio corresponding to different input intensities. Consequently, we are able to determine the tip sample separation.

Since our scanning probe microscope operates in intermittent contact mode, one expects a minimum tip sample separation close to zero. Using the procedure outlined above, we obtain $z = 74 \pm 1$ nm which represents an effective tip-sample separation due to the slanted geometry

of our probe. Using this value in Eq. (4), we generated maps of the measured gradients of the optically induced force $\partial F_{opt} / \partial z$. As can be seen in Figure 5-2, this gradient is negative and is of the order of $10^{-7} N/m$, which is comparable to the retarded van der Waals force experienced by our tip when it is tens of nanometers away from the surface.

Figure 5-2 Gradient of the optical force extracted from the perceived topography shown in Fig. 5-1 when (a) 24 and (b) 16 mW power is coupled into a single mode fiber.

In order to further assess these results, we estimated the optical forces by performing numerical simulations using the Finite Element Method (Comsol Multiphysics 3.5a), which account for the complex three-dimensional behavior of the optical forces near the surface and its interaction with a realistic tip. To provide an accurate representation of the tip used in the experiments, the probe was modeled as a hollow cone with an opening angle of $\approx 9^\circ$ and an aperture consisting of a 100 nm diameter core and a 100 nm thick dielectric cladding with a refractive index of 1.5. The cone axis was tilted by 30° with respect to the normal to the scanning plane and the length of the simulated probe was $\sim 4 \mu m$, which is limited only by the computational capabilities available (Intel Core2 Duo, 3GHz, 8GB of RAM). We expect this to

effectively model an infinite tip for the scan points in the vicinity of the fiber core. The field emerging from a single mode fiber with a $3.5 \mu m$ effective mode diameter was modeled as a Gaussian beam with its waist located at the end of the fiber. An example of the field distribution in the cross-section of the probe is shown in Figure 5-3(a). Using the calculated field distribution and the Maxwell stress tensor representation [101], we then evaluated the optical force acting on the scanning probe. Note that with this approach, all the components of optical force, gradient force, radiation pressure, force due to phase gradient [102], are taken into account.

When the probe is located $1 \mu m$ past the fiber core as in Figure 5-3(a), i.e. approximately where the arrow is pointing to in the inset of Figure 5-1(b), we find that F_{opt} is on the order of $10^{-11} N$ for a $24 mW$ illuminating beam, which is comparable with typical surface forces. Calculations also show that the gradient of the optical force is nearly constant over the range of the tip oscillation considered and has a value of $-1.6 \cdot 10^{-7} N/m$, which is similar to the gradient of the dispersion force at a distance $z \approx 65 nm$. We note that even though the field distribution varies laterally over the scan area, at each point in the scan the gradient may be assumed constant in the longitudinal direction over the range of the tip oscillation amplitude.

We also note that the electromagnetic interaction between the probe and fiber itself may, in principle, affect the field distribution and, consequently, the interaction forces. However, our systematic evaluations indicate that the effect of this so-called optical binding force [62,68,103] that establishes between dielectric bodies irradiated by a common electromagnetic field is about two orders of magnitude weaker than the other optical forces near the surface and almost vanishes when the tip-sample separation is $\sim 250 nm$. Because the frequency shift is

determined by the average force acting on the tip during one oscillation cycle [18,100], we expect that the optical binding force provides only a small contribution to the overall frequency shift when averaged over the large oscillation amplitude ($\sim 90 \text{ nm}$) used in our experiment. Thus, to first order, the influence of binding forces can be neglected.

Figure 5-3 (a) Simulated probe geometry together with electric field distribution and (b) the calculated (solid line) and measured (symbols) gradient of the optical force acting on a probe scanning over the core of fiber along the direction indicated in the inset of Fig.1b. The error bars account for the errors associated with estimating z and A along with the noise in the topography maps.

We can now quantitatively compare the values of the optical force gradient obtained experimentally with the results of the full electromagnetic calculation. The results are presented in Figure 5-3(b) for different positions of the probe scanned in the direction indicated by the arrow in the inset of Figure 5-1(b). As can be seen, there is an excellent quantitative agreement between the simulated and measured cases. We note that the main sources of experimental errors are due to the accuracy in determining z and A as well as the noise in the topography signals.

5.5 Optical forces in evanescent fields

In the proof of concept experiment described above we demonstrated that optical forces can be measured with NSOM probes scanned through a complex optical field. However, in the example illustrated in Figure 5-3(a), the main contributions are due to homogeneous field components. To investigate the possibility of measuring optically induced forces in arbitrary fields, we conducted a second experiment where the optical field is evanescent and is thus measured in a more traditional scanning probe configuration. Here we use a standard pulled fiber NSOM probe with a 100 nm aperture which is coated with a 10 nm Cr adhesion layer and a 250 nm thick gold coating. This tip is scanned over the focused spot of a 532 nm laser illuminating a triangular prism in a total internal reflection (TIR) configuration.

To compute the optically induced force, the analysis described above may be repeated except that now Eq. (5.4) should be replaced with

$$\left[M_1^{0.5}(-2\alpha A) - M_2^{1.5}(-2\alpha A) \right] = B_L R \left[\frac{1}{z^n} F_W^n - \frac{1}{(z + \delta z)^n} F_{W-opt}^n \right] \quad (5.5)$$

to account for the exponential decay of the radiation from the surface [100]. $M_a^b(z)$ in Eq. (5.5) is the so-called Kummer's function [100], F_0 is the force at the surface of the prism, $\alpha = 2k\sqrt{n^2 \sin^2 \theta_i - 1}$ is the decay constant, and θ_i is the angle of incidence at the top surface of the prism. From the experimental measurements we find $\alpha = 0.00805 \text{ nm}^{-1}$, which correlates very well with $\alpha = 0.00836 \text{ nm}^{-1}$ obtained from the above formula for $\theta_i \approx 45^\circ$ used in experiment. This confirms the electromagnetic origin of measured forces. The constants B_L and n in Eq. (5.5) are estimated based on the results in Ref. [104] where the force between

an Au sphere in air and a silica plane is calculated using Lifshitz theory. We note that, as can be seen from Eq. (5.5), in the case of evanescent fields the optical force itself is measured rather than its gradient. This may be simply understood by realizing that the derivative of an exponential (evanescent decay) is also an exponential.

In Figure 5-4(a) we show the measured topography when $\sim 75 \text{ mW}$ of laser light was focused onto the prism surface at the angle $\theta_i \approx 45^\circ$ resulting in a maximum intensity of $\approx 5 \text{ mW}/\mu\text{m}^2$ at the surface. The spot size of the illumination (FWHM) is $\sim 13.2 \mu\text{m}$ along the long axis. One can clearly see the influence of the optically induced force, even for the moderate intensity used here. Repeating the scan with the light turned off (not shown) revealed a flat surface with a rms roughness less than 1 nm . The optically induced force at the prism surface, F_0 , as evaluated from Eq. (5.5) is shown in Figure 5-4(b).

Figure 5-4 (a) Topography and (b) extracted optically induced force at the surface of a prism due to a focused laser beam illuminated in a TIR condition.

We can compare again the experimental results with the estimations of our full electromagnetic calculations. According to the probe's specifications, we model it as a cone with a 30° angle at the apex with the axis of the cone slanted with respect to the normal to the surface by 15° . A $1\ \mu m$ long portion of the probe was modeled; because the optical field rapidly decreases away from the surface, a $1\ \mu m$ long portion of the probe is sufficient to obtain a precise force estimation. The optically induced forces acting on the probe were calculated using Maxwell's stress tensor. We found that the force at the center of the illumination spot is $F_o^{theor} \approx 1.6\ pN$. This value is somewhat smaller the experimentally measured force of $F_o^{exp} \approx 6 \pm 2\ pN$ extracted using the procedure outlined before. Note that, because practically only the very tip of the probe interacts with the optical radiation, this force simply scales with intensity across the illumination spot. One of the reasons for the difference between the values of the forces calculated numerically and measured experimentally may be our approximation of the NSOM probe as a solid Au sphere for the calculation of the van der Waals forces. Another possible aspect could be the influence of thermal (photophoretic) forces that can arise because of resistive heating in the metal coating on the probe. In order to assess the potential role of thermal effects a transient heat transfer analysis was performed using COMSOL. Calculations show that the stationary regime for the temperature distribution at the tip is achieved within $2\ ms$ after exposure to the illumination. The temperature at the tip rises on average by $30\ K$ and oscillates with amplitude of about $9\ K$ following the mechanical oscillations of the probe. As our measurement technique is sensitive not to the absolute magnitude of the force but to the change

of the force over the probe's oscillation cycle, these small temperature oscillations may have a minor contribution to the measured force F_0^{exp} .

5.6 Applications

In this Chapter we introduced a new way to sense properties of both propagating and evanescent electromagnetic fields. By measuring the mechanical action exerted on a standard scanning probe, one can acquire maps of the optically induced topography from which the gradient of the optically induced force can be evaluated. Notably, if such maps are collected at different intensities, the tip sample separation can be determined without employing additional hardware. The approach permits quantitative measurements as demonstrated by the good agreement with full electrodynamics calculations.

In the case of the detection of propagating fields, we demonstrated a new modality to measure optically induced forces, which allows for the direct detection of the gradient of the force. In addition, we have shown that the influence of optical forces is significant for standard NSOM probes even when illuminated by evanescent waves at moderate intensities. Therefore, the effect of optical forces should be carefully considered in the practice of standard NSOM measurements.

Because the optical field is not actually coupled into the probe, our measurement has a unique characteristic: the spatial resolution is not limited by the size of the aperture one can practically use, an important feature present also in s-NSOM measurements. However, as opposed to s-NSOM, we obtain this advantage without using any additional optics, photon detection equipment, and the signal processing necessary to suppress the background scattering.

In particular, the fact that a photon detector is not required opens up the possibility to measure radiation that is not readily detectable with standard detectors, e.g., far infrared and THz radiation.

While the measurements presented in this paper were conducted with the feedback on such that the information about optical force was embedded in the topography signal, we could have alternatively maintained a constant tip-sample separation and used the phase signal in order to extract the same information. Such an extension is straightforward and may be used to determine the optical force while the probe remains close to the surface. Moreover, a similar measurement and data analysis can be performed for fields in free space without the presence of a material interface.

6 DISCRIMINATION OF FIELD COMPONENTS IN NEAR-FIELD SCANNING OPTICAL MICROSCOPY

6.1 Introduction

The previous two chapters qualitatively and quantitatively demonstrated the effect of the gradient of optically induced force on the resonance frequency of the tuning fork and, therefore, on its feedback. This caused spurious features in the topography due to this additional contribution of the optically-induced topography. In the previous chapter we demonstrated how these effects can be used to quantify the strength of the OIF gradient by using another, quantitatively known force as a yardstick. In our particular case, this force reference was provided by the van der Waals force on a fused silica surface. In the same time, the superposition of these two different force fields allowed us to determine the actual tip-sample separation at closest approach in a relatively simple manner. In this chapter we discuss an additional application for the measurement of the gradient of the optically induced force. Specifically, we will show that the OIF effects provide a complementary measurement of the complex local field distribution as compared to conventional a-NSOM measurements.

The measurement of light typically employs a photon-to-electron converting device, i.e. a photodetector that effectively measures the light intensity. Specific components of the optical field can be detected only when some combination of waveplates and polarizers are added in front of the detector to isolate the particular field component of interest that is then measured with a photodetector. Imaging systems rely on this procedure to record spatially-resolved light distributions, but their resolution is usually diffraction limited. This drawback can be overcome

using, for instance, scanning approaches where the spatial resolution is determined by the volume of interaction between the optical radiation and a small probe. Due to the nature of this complex interaction, the measurement is sensitive to different field components. For example, depending on the exact probe, either electric or magnetic fields in the plane of the probe aperture may be predominately collected in near-field scanning optical microscopy (NSOM) [105,106].

It has been demonstrated in Chapter 4 that using standard NSOM and atomic force microscope (AFM) probes, light may be measured not only by counting photons in the usual manner but also by detecting the optically induced forces, which manifest themselves as an additional contribution to topography [23]. This sort of measurement is typically described as an alternative measurement to circumvent the photon counting [99]. In this chapter we will demonstrate that, in fact, it is a complementary measurement modality that provides information not available in standard NSOM measurements.

6.2 Experiment

To demonstrate the complementary nature of the NSOM signal and optically induced force (OIF) measurements, we used a Nanonics MultiView 4000 NSOM to perform a standard collection mode NSOM scan of a standing wave pattern formed by counter-propagating evanescent waves. The evanescent waves were generated by two slightly focused 532 *nm* laser light beams incident on the surface of a prism ($n=1.5$) past the critical angle ($\theta_3 = 51^\circ$), as shown in Figure 6-1. The probe was a standard Au coated pulled silica fiber probe. The images for transverse electric (TE) and transverse magnetic (TM) polarizations are shown in Figure 6-2. A standard collection mode scan with the probe operated in an intermittent contact mode was

used. Interference fringes are clearly observed not only in the NSOM images but also in the topography, which is proportional to the gradient of force $\partial F_z / \partial z$ [23]. Topography scans taken with the light off (not shown) reveal that the prism surface is flat except for the few small features observed in the upper right corner of Figure 6-2 (a) and (b).

Figure 6-1: Schematic of experiment. An interference pattern is generated on the surface of a prism using two focused laser beams. A standard NSOM tip is scanned over this focused spot to generate a map of the topography for both TE and TM illumination..

To aid with the visualization, we averaged the data along each fringe, as shown in Figure 6-3. The TE and TM polarization data sets were aligned with respect to one another using the small features present in the upper right corner of the topography images. As can be seen in Figure 6-3, in the case of TE illumination, the NSOM intensity and the topography fringes are relatively well aligned with each other. The small overall decay is simply due to the fact that the location of the measurement was not exactly at the center of the counter propagating beams. In

the case of TM illumination however, we note a strikingly different, anti-phased behavior of the two sets of fringes.

Figure 6-2: Topography (force gradient) when (a) TE and (b) TM polarized light is incident on the prism surface; (c) and (d) are the same for the NSOM signal.

As explained below, the fact that the fringes are aligned for TE polarization and anti-aligned for TM polarization case demonstrates that the force and NSOM measurements provide complementary measures of the local field distribution.

Careful study of Figure 6-3 reveals that the fringe spacing is $226 \pm 3 \text{ nm}$, which compares favorably with the theoretically expected 228 nm fringe spacing. The TE NSOM signal is shifted $\sim 14 \text{ nm}$ behind the TE height signal (modulo 226 nm) and the TM NSOM signal is shifted $\sim 92 \text{ nm}$ behind of the TM height signal, or $\sim 19 \text{ nm}$ ahead of anti-phase alignment. One of the possible explanations of these shifts is that the topography and the NSOM signals originate from the different locations of the aperture probe. However, this cannot explain different magnitudes and directions of these shifts. As we will justify at the end of this chapter, this may be explained by the detection of both electric and magnetic field components by our probe.

6.3 Simple Probe Model

To explain the results, we need to model the collected NSOM signal and optically induced force on a metal coated NSOM probe. In accordance with our experimental configuration, the electric field at the surface of the prism is given by

$$\begin{aligned} E_x &= (2i/n_{21}) t_{21, TM} t_{43, TM} E_{0, TM} \sqrt{\sin^2(\theta_3) - n_{21}^2} \cos(\gamma x) \exp(-\beta z) \\ E_y &= 2t_{21, TE} t_{43, TE} E_{0, TE} \cos(\gamma x) \exp(-\beta z) \\ E_z &= (-2i/n_{21}) t_{21, TM} t_{43, TM} E_{0, TM} \sin(\theta_3) \sin(\gamma x) \exp(-\beta z) \end{aligned} \quad (6.1)$$

where $n_{21} = n_2/n_1$. The transmission coefficients, $t_{21, Tx}$ (see Figure 6-1), may be found in any introductory textbook on the subject; see for example [107]. Likewise, expressions for $t_{43, Tx}$ (Figure 6-1) as well as the wavevectors β and γ may be found in [89]. Similar equations may also be derived for the magnetic fields. However, because of the plane wave illumination, the

magnetic field components are all in phase with the electric field components and $B_z/B_x = -E_z/E_x$.

Having described the complex field at the surface of the prism, we develop models for the probe NSOM and force signal collection. For reasons we justify at the end, we consider our probe to be primarily sensitive to the electric fields. Because the tip's aperture is at an angle θ_4 with respect to the surface, the electric field components in the plane of the aperture are $E_{tip-x'} = E_x$ and $E_{tip-y'} = E_y \cos(\theta_4) + E_z \sin(\theta_4)$. Thus, the collected NSOM intensities for TE and TM polarization are

$$\begin{aligned} I_{NSOM_TE} &\propto |E_y|^2 \cos^2(\theta_4) \\ I_{NSOM_TM} &\propto [|E_x|^2 + |E_z|^2 \sin^2(\theta_4)]. \end{aligned} \quad (6.2)$$

Because the long axis of our probe is at a finite angle with respect to the surface normal, the metal coating is thick (~ 250 nm), and because we are using evanescent excitation, we expect the optically induced forces to predominately affect the gold coating. Therefore to compute the OIF, we model the probe as a gold prolate spheroid tilted at an angle θ_4 with respect to the normal to the prism surface. We use this model because it incorporates the essential features of the actual probe geometry while retaining an analytical solution. The induced probe dipole moments for TE and TM incident polarizations are given by

$$\begin{aligned} p_{TE} &= \epsilon_0 \begin{bmatrix} \hat{y}(\alpha_y \cos^2(\theta_4) + \alpha_z \sin^2(\theta_4)) E_y + \\ \hat{z} \cos(\theta_4) \sin(\theta_4) (\alpha_y - \alpha_z) E_y \end{bmatrix} \\ p_{TM} &= \epsilon_0 \begin{bmatrix} \hat{x} \alpha_x E_x + \hat{y} \cos(\theta_4) \sin(\theta_4) (\alpha_y - \alpha_z) E_z + \\ \hat{z} (\alpha_z \cos^2(\theta_4) + \alpha_y \sin^2(\theta_4)) E_z \end{bmatrix} \end{aligned} \quad (6.3)$$

where $\alpha_{x,y,z}$ are the polarizabilities along the major and minor axes of a spheroid, as given in [108].

Using Eq. (6.3) we evaluated the force induced on an equivalent anisotropic electric dipole [89], and found that for evanescent wave excitation, the two components of the force gradient are given by:

$$\begin{aligned} \left(\frac{\partial F_z}{\partial z} \right)_{TE} &= 2\beta^2 \varepsilon_0 |\mathbf{E}_y|^2 \operatorname{Re}(\alpha_y \cos^2(\theta_4) + \alpha_z \sin^2(\theta_4)) \\ \left(\frac{\partial F_z}{\partial z} \right)_{TM} &= 2\beta^2 \varepsilon_0 |\mathbf{E}_x|^2 \operatorname{Re}(\alpha_x) + \\ &\quad |\mathbf{E}_z|^2 \operatorname{Re}(\alpha_z \cos^2(\theta_4) + \alpha_y \sin^2(\theta_4)) \end{aligned} \quad (6.4)$$

A positive gradient corresponds to the tip retracting while a negative force gradient corresponds to the tip approaching the sample surface [23].

Figure 6-3: Averaged height of optically induced topography (solid red lines) and NSOM signal (dashed blue lines) for (a) TE polarization and (b) TM polarization.

We are now in a position to compare the NSOM and force gradient signals given in Eqs. (6.2) and (6.4), respectively. For TE illumination both signals are proportional to $|E_y|^2$ and thus

are linearly dependent. However, for TM polarization, the NSOM and force signals depend on different combinations of $|E_x|^2$, $|E_z|^2$ and, therefore, carry complementary information about composition of the near-field distribution. In general, this information will depend on the particular probe parameters.

Using Eqs. (6.2) and (6.4), we calculate the expected NSOM and topography signals; typical results are shown in Figure 6-4. Here we model our probe as a spheroid with an aspect ratio of $a:b:c = 1:1:2.5$ tilted at an angle $\theta_4 = 30^\circ$. We observe excellent qualitative agreement between our experimental results and our model, even for the relatively simple model employed here [109].

The fringe positioning can be understood by examining Eq. (1) where one can see that I_x and I_y are proportional to $\cos^2(\gamma x)$ whereas I_z is proportional to $\sin^2(\gamma x)$. Because θ_4 controls the relative coupling between I_x and I_z for I_{NSOM_TM} , this determines whether the NSOM fringes are aligned or anti-aligned. Because I_z is much stronger than I_x , it does not take a high tip angle for I_z to overcome I_x .

The OIF fringes are aligned because of the effect the aspect ratio (AR) has on the sign of the spheroid polarizability components. Specifically p_z is negative for $AR > \sim 2.2$. Thus, the $\Re(p_z E_z^*)$ contribution to $\partial F_z / \partial z$ is repulsive whereas $\Re(p_x E_x^*)$ and $\Re(p_y E_y^*)$ are both attractive. Therefore the fringe maxima from all three contributions are aligned.

As stated earlier, we assumed that the NSOM probe is sensitive to only the electric field. The magnetic component of the evanescent field can, in principle, also contribute to the detected signal [100]. For instance, in TM polarization the probe would be sensitive to the H_x component

while for TE polarization the probe would detect the H_y and H_z components with $|H_z| \gg |H_y|$. As H_x is in phase with E_x and E_y , the magnetic fields will then tend to bring I_{NSOM_TM} in phase with the topography signal. On the contrary, in TE polarization H_z will tend to shift the detected I_{NSOM_TE} out of phase with the induced topography. Because we do not observe this in our experiment, this means that, for our probes, coupling to the electric fields dominates the NSOM response. However, the presence of such a magnetic field contribution is able to explain the different directions of the relative shift between the NSOM and topography signals for TE and TM illumination.

Figure 6-4: Calculated NSOM signals (dashed blue lines) and gradient of the force signals, which are nonlinearly related to the topography (solid red lines) for (a) TE and (b) TM polarization.

6.4 Conclusion

In this Chapter we demonstrated that the measurements of the optical intensity using an aperture NSOM probe and optically induced force are complementary. By measuring with a photodetector the standard NSOM optical signal while simultaneously detecting the force gradients that are optically induced on the same probe, one can access different components of

the complex 3-D field distribution. Most importantly, this additional information is acquired without increasing the complexity of the measurement or of the probe. In conclusion, one can state that understanding the effect of optically induced forces not only assists in the correct interpretation of NSOM data [9], but also opens up the possibility to simultaneously acquire additional samplings of the complex 3-D field distribution incident upon the probe without increasing the measurement complexity. With a well characterized probe, it should then be possible to separate out the different components of the electromagnetic field.

7 MULTIFREQUENCY NEAR-FIELD SCANNING OPTICAL MICROSCOPY (MF-NSOM)

7.1 Introduction

In the previous chapters, we demonstrated the possibility and discussed the implications of using OIF to detect light at levels consistent with traditional NSOM measurements. In those experiments, the optical field used to illuminate the probe did not vary in time. In this case, we practically measured the OIF gradient, which acted to shift the probe resonance frequency and/or damping. In this chapter we explore the possibility to directly drive the probe by modulating the optical field in a manner analogous to multi-frequency atomic force microscopy (MF-AFM) [108,109]. The basic MF-AFM modality involves the electrical excitation of an AFM probe at two different frequencies. The first frequency is used to regulate the tip-sample distance as in standard AFM operation. The second frequency, usually driven with much smaller amplitude, is used to probe additional properties of the sample. The primary advantage of utilizing multiple frequencies is that each frequency contains a unique, albeit complicated, sampling of the tip-sample interaction force. Adding this extra information allows to separate the topography from the forces acting on the probe, separate the short and long range forces acting on the probe, locally characterize the tip-sample interaction to simultaneously extract several material properties or perform a real time determination of the cantilever properties [111–113]. One interesting feature of multi-frequency imaging is the mixing that occurs between the different frequencies due to the fact that the probe is oscillating in a nonlinear surface force interaction potential. One can take advantage of the mixing to, for example, image subsurface

features by driving the probe at one frequency and the sample at another and measuring the response at mixing frequencies [111].

Here we demonstrate that the multi-frequency approach can be applied to NSOM by modulating the light excitation and, consequently, driving optically the probe's oscillations. In analogy to MF-AFM, this allows us to simultaneously measure different surface properties. Specifically, the topography is recorded as in standard AFM by applying a feedback loop to the signal from the tuning fork and then using a lock-in to measure the signal at the frequency of electrical excitation. At the same time, the optical force due to the near-field distribution is measured by modulating the light at another frequency and using a second lock-in detection. Hence, this operation modality replaces the small amplitude electrical excitation of the probe with the small amplitude optical excitation; therefore we refer to this technique as multi-frequency near-field scanning optical microscopy (MF-NSOM). This is in contrast to the measurements shown in the previous chapters where the gradient of the optically induced force was mixed with surface force gradient in the topography signal [99,110,114].

By detecting the electromagnetic field by the force it exerts on the probe, we are able to perform the detection of light in the near-field for a broad range of wavelengths. Somewhat related concepts have been approached before. For example, forming a Schottky diode on the probe [115,116], utilizing the piezoresistive effect [117], using the image force [118] or taking advantage of surface photovoltage induced forces acting on a semiconductor probe [54,56,119] have also relied on measuring the optical signal or optically induced forces locally with the probe rather than using the probe to scatter the light into the far field. We add to these reports in a number of ways. Most importantly, we demonstrate the measurement of the magnitude and

phase of optically-induced forces using tuning fork-based systems that utilize all dielectric probes. This eliminates or at least severely diminishes the possibility of thermally-induced effects. We also show that the effect of surface forces cannot be completely isolated by simply modulating the intensity of light at high frequency. Rather, the surface- and light-induced influences are coupled by the force gradients acting on the probe. This mixing occurs because the probe oscillates in a mixed nonlinear interaction potential.

7.2 Demonstration of MF-NSOM

In the following we will demonstrate that tuning fork based systems can be used to measure high-resolution images of near-field optical forces. In order to achieve the desirable sensitivity, systems based on tuning forks need to be operated close to their mechanical resonances, a requirement that is not so stringent in the case of cantilever based systems. The advantage of the tuning fork, however, is an efficient suppression of all off-resonance contributions, which allows for a more controllable excitation of the probe. In the present experiments, we used a tuning fork based NSOM operated at the lowest order resonance (typically $\sim 30 - 40 \text{ kHz}$) for feedback control as in standard AFM and at the next higher order resonance (typically $\sim 180 - 250 \text{ kHz}$) for optically induced excitation.

The physical origin of the measured signals can be described by considering the NSOM probe as a damped oscillator [120,121]. This model can account for the influence of both the external force, through direct excitation of oscillations, and its gradient, through changes in resonator's properties. Indeed, while the topography (i.e. the gradient of the surface forces) and the optical force are measured separately at two different frequencies, they are coupled by the gradients of both surface and optically induced forces which modify the local effective tuning

fork resonator properties. As we will show, it is possible however to extract the optically induced force from the measured amplitude and phase at the optical modulation frequency.

To conceptually understand the origin of the optically induced force effects, we model our tuning fork based NSOM system using the well-known mass on a spring model [120,121]. Though a more rigorous model with two coupled equations of motion, one for each arm of the tuning fork, would more rigorously describe the physics, this simple model is sufficient to describe the system dynamics [122]. In this case the equation of motion for an oscillator of mass m is

$$m\left(\ddot{a} + \frac{\omega_0}{Q}\dot{a} + \omega_0^2 a\right) = F_{drive}(t) + F_{surface}(x, y, z) + F_{optical}(x, y, z) \quad (7.1)$$

where a is the location of the probe during its oscillation cycle along the direction z of its oscillation, Q is the quality factor, ω_0 is the eigenfrequency of the oscillator, F_{drive} is the electrical driving force which is used for feedback control, $F_{surface}$ denotes surface forces, while $F_{optical}$ encompasses all of the optically induced forces. When using the feedback to perform a constant gap mode scan, the tuning fork is electrically driven by $F_{drive} = F_d \exp(i\omega_1 t)$ and then lock-in detection is used to read the reflected signal from the oscillating tuning fork: $a(t) = A_1 \exp(i\omega_1 t + i\phi_1)$. We assume that the gradients of both the surface and the optically-induced forces are small over the oscillation cycle so we may use a two term Taylor expansion for $F_{surface}$ and $F_{optical}$. This assumption is made only for the sake of clarity; the physics remains essentially the same even in cases where this assumption is not valid [100].

In these conditions, $F_{surface}(x, y, z) = F_{surf}(x, y, z) + (\partial F_{surf}(x, y, z)/\partial a)a$, $F_{optical}(x, y, z) = F_{opt}(x, y, z) + (\partial F_{opt}(x, y, z)/\partial a)a$, and using these expressions in Eq. (7.1), one obtains

$$\left(\omega_0^2 - \omega_1^2 + i \frac{\omega_0}{Q} \omega_1 - \frac{1}{m} \frac{\partial (F_{surf}(x, y, z) + F_{opt}(x, y, z))}{\partial a} \right) A_1 \exp(i\phi_1) = \frac{F_d}{m} \quad (7.2)$$

where we note that a is always along the z direction in our treatment.

As can be seen, the surface and the optically induced force gradients act to shift the resonance frequency of the probe, and therefore they change the amplitude and phase detected with the lock-in set at ω_1 .

Careful inspection of Eq. (7.1) however reveals a second possibility for detecting optical force effects: direct measurement of the force via modulating the light and using an additional lock-in to measure the signal at the optical modulation frequency. In this case we have

$$\begin{aligned} m \left(\ddot{a}_1 + \frac{\omega_{01}}{Q_1} \dot{a}_1 + \omega_{01}^2 a_1 \right) &= F_{drive}(t) + F_{surface}(x, y, z) + F_{optical}(x, y, z, t) \\ m \left(\ddot{a}_2 + \frac{\omega_{02}}{Q_2} \dot{a}_2 + \omega_{02}^2 a_2 \right) &= F_{drive}(t) + F_{surface}(x, y, z) + F_{optical}(x, y, z, t) \end{aligned} \quad (7.3)$$

where the displacement is now $a = a_1(t) + a_2(t) + O(\varepsilon) \approx A_1 \exp(i\omega_1 t + i\phi_1) + A_2 \exp(i\omega_2 t + i\phi_2)$ and

$F_{optical} = F_{opt_avg} + \frac{\partial F_{opt_avg}}{\partial a} a + F_{opt} \exp(i\omega_2 t) + \frac{\partial F_{opt}}{\partial a} a [\exp(i\omega_2 t)]$. The total optical force contains four terms because the light intensity is modulated between zero and a finite maximum value and, thus, the average intensity is non-zero. The first and last terms in $F_{optical}$ do not

influence the measurements at the two lock-in frequencies, ω_1 and ω_2 . At these frequencies the corresponding equations of motion read

$$\begin{aligned} \left(\omega_{01}^2 - \omega_1^2 + i \frac{\omega_{01}}{Q_1} \omega_1 - \frac{1}{m} \frac{\partial (F_{surf}(x, y; z) + F_{opt_avg}(x, y; z))}{\partial a_1} \right) A_1 \exp(i\phi_1) &= \frac{F_d}{m} \\ \left(\omega_{02}^2 - \omega_2^2 + i \frac{\omega_{02}}{Q_2} \omega_2 - \frac{1}{m} \frac{\partial (F_{surf}(x, y; z) + F_{opt_avg}(x, y; z))}{\partial a_2} \right) A_2 \exp(i\phi_2) &= \frac{F_{opt}}{m} \end{aligned} \quad (7.4)$$

where ω_{01} and ω_{02} are the closest eigenfrequencies to ω_1 and ω_2 , respectively. Eq. (7.4)(b) is essentially the same as Eq. (7.1) where $\partial F_{opt}/\partial a$ has been replaced by $\partial F_{opt_avg}/\partial a$. Thus, even though we modulate the light, the gradient of the optical force still affects the topography measurement due to the fact that the average intensity is always finite. Also, as can be seen in Eq. (7.4), the topographical and optical signals are not entirely decoupled as $\partial F_{surf}/\partial a$ and $\partial F_{opt}/\partial a$ enter both equations. Below we will discuss the possibility to decouple these influences.

In our experiments, we use a lock-in amplifier to measure the amplitude and phase of the signal excited on the tuning fork at the optical modulation frequency. Using the mass on a spring model, these are found to be

$$\begin{aligned} A_2 &= \frac{F_{opt}}{m} \frac{1}{\sqrt{\left(\omega_{02}^2 - \omega_2^2 - \frac{1}{m} \frac{\partial (F_{surf}(x, y; z) + F_{opt_avg}(x, y; z))}{\partial a_2} \right)^2 + \left(\frac{\omega_{02}}{Q_2} \omega_2 \right)^2}} \\ \phi_2 &= \tan^{-1} \left(\frac{(\omega_{02} \omega_2)/Q_2}{\omega_{02}^2 - \omega_2^2 - \frac{1}{m} \frac{\partial (F_{surf}(x, y; z) + F_{opt_avg}(x, y; z))}{\partial a_2}} \right) \end{aligned} \quad (7.5)$$

These equations have a few implications. First, we only expect to measure a phase change if there is a measurable change in the gradient of the sum of the surface and/or optically induced forces. Second, by measuring both the amplitude and the phase, we should be able to separate the effects of optically induced force from the combined influence of the surface and optically induced force gradients. Decoupling the optically and surface induced force gradients can be done, but it requires subtracting two topography signals, recorded at the frequency of the electrical modulation with and without illumination, as we have shown in a previous publication [110].

It is known that a tuning fork based NSOM is more exactly modeled as two coupled oscillators which are unbalanced due to the additional mass (i.e. the probe) plus additional external gradients acting on one arm [119,120]. However, these effects can be neglected here because the influence of the additional mass is constant and the force gradients arising from optically and surface induced forces (averaged over the relatively large oscillation amplitudes of tens of nm) are expected to negligibly affect the balance of the prongs. Therefore the simple damped oscillator formalism provides an accurate description of the physics of our system.

To demonstrate the feasibility of MF-NSOM we performed a proof of concept experiment where we illuminated an NSOM tip from beneath with 635 nm laser light coupled into a single mode fiber as shown in Figure 7-1. To demonstrate the possibility to optically drive the oscillations of the tuning fork, the laser emission was intensity modulated while the peak average intensity at the fiber face was maintained at a moderate level of $0.36 \text{ mW}/\mu\text{m}^2$. The probe was placed over the core of the fiber and then retracted $2 \mu\text{m}$. The modulation frequency of the laser diode was scanned across the first two resonances of the tuning fork and the signal

from the tuning fork was demodulated at the optical modulation frequency using a lock-in amplifier. This procedure provided a direct measure of the piezoelectric signal generated by the optical force onto the tuning fork. Note that the electrical excitation of the quartz tuning fork was moved far off resonance to prevent it from interfering with the measurement of the optically induced forces.

Figure 7-1: Schematic of multi-frequency NSOM setup used in the proof-of-concept experiment.

The results of these scans, along with the corresponding standard electrical excitation results, are shown in Figure 7-2. As can be seen, a measurable signal is induced by modulating the optical excitation with the results of optical and electrical excitation largely coinciding. Careful inspection of the data reveals that at the higher-order resonance the response is stronger for electrical excitation, while for the low frequency resonance the situation is reversed. This comes about because of the different excitation conditions. When driven electrically, the probe is acted on at the base of both arms of the tuning fork while the optical excitation is locally applied only on the bottom arm of the fork. The observed resonances correspond to the two lowest order anti-symmetric in-plane eigenmodes. Because of the local nature of the optical excitation, the

optical modulation excites the lowest order resonance more effectively than the higher one leading to higher oscillation amplitude as seen in Figure 7-2(a). By contrast, the electrical signal is stronger due to the higher sensitivity of the harmonic due to its lower effective spring constant [122]. The asymmetry in the amplitude traces is due to a parasitic capacitance present in the tuning fork [120]. The small frequency shift between the optical and electrical excitation observed in Figure 7-2 may be due to drift in the tuning fork response, inaccuracies in the extraction of the points for the electrical excitation, differences in the calibration of the different lock-ins used for the two measurements or a force gradient induced shift in the resonance frequency of the tuning fork.

Figure 7-2: Magnitude and phase of the current induced on the tuning fork as a result of optically induced excitation (blue solid curves) and electrical driving (red dashed curves). (a) and (b) are the magnitude and phase at the lowest resonance while (c) and (d) are the magnitude and phase at the next higher resonance.

7.3 Characteristics of optical force imaging

To demonstrate the imaging capability of optical force detection, we present a series of measurements taken with an uncoated NSOM probe over the face of a single mode fiber with 1310 *nm* laser diode light coupled out as well as a 635 *nm* laser diode coupled to the same type of fiber. This second case demonstrates the complex amplitude and phase responses of the

probe at the optical modulation frequency that may be observed when imaging a complex spatial distribution realized by the slightly multimode nature of the fiber. These additional examples demonstrate some of the key characteristics of optical force imaging.

In Figure 7-3 we show the spectra of the higher order resonance for the probe used in Figure 7-4 and Figure 7-5. The red dashed lines indicate the three frequencies used in the three leftmost columns of Figure 7-4 while the blue dash-dot line shows the frequency used in Figure 7-5.

Figure 7-3: Frequency sweep of higher order resonance of uncoated NSOM probe used for scans. Red dashed lines indicate frequencies used for left three columns of Figure 7-4. The blue dash-dot line indicates the frequency used in Figure 7-5.

Reading down each column in Figure 7-4, we see the topography followed by the NSOM signal and then the amplitude and phase of the detected signal related to optically induced force. The signal is collected at the light modulation frequency excited on the higher order resonance of the NSOM's tuning fork while the uncoated NSOM probe is scanned over the core of a single

mode fiber with 1310 *nm* light coupled out. The left three columns show the results somewhat below, at and somewhat above resonance, as shown in Figure 7-3. In Figure 7-4, we see the expected result that the amplitude is a maximum on resonance and tails off away from the resonance while the phase decreases as the frequency increases, just as it does around resonance in Figure 7-4. Two more plots farther above and below resonance (not shown) further show this trend.

The NSOM signal is distorted with a “comet tail” in the upper left corner. This distortion occurs because the probe is oriented such that it passes from the lower right to the upper left corner. Because the probe is uncoated and because light emerging from the fiber is mainly propagating, light is able to couple in farther up the probe. The large signal over the core is due to optical binding between the probe and its image. The “comet tail” conversely is due to radiation pressure acting farther up the shaft of the probe. This is confirmed by Figure 7-4(s-t) which shows the collected signal on resonance with the probe retracted many microns. Here we see that the spot over the core is gone but the comet tail remains.

The topographies shown in the three leftmost columns contain two dips which are not present when the light is off. This is the “optically induced topography” resulting from the gradient of the average light intensity, as expected from Eq. (7.4). The presence of the two dips in these observations is due to the specific way light coupled to this particular probe.

Figure 7-4: Reading down the columns, each shows the topography of the face of a single mode fiber followed by the NSOM signal and amplitude and phase respectively measured at the optical modulation frequency for an uncoated NSOM probe scanning over the fiber's face with 1310 *nm* light coupled out. The left three columns correspond to frequencies somewhat below ($f = 241.37 \text{ kHz}$), at ($f = 241.59 \text{ kHz}$) and somewhat above ($f = 241.94 \text{ kHz}$) the higher order resonance. The fourth column shows data collected with the light turned off. The rightmost column shows data at an excitation frequency of $f = 241.37 \text{ kHz}$ collected when the probe is many microns above the surface.

In a subsequent experiment we replaced the 1310 *nm* laser diode with a 635 *nm* laser diode such that the light coupled out is slightly multimode. The end face of the fiber was scanned with the same uncoated NSOM probe while the light was modulated with a frequency slightly above the higher order resonance ($f = 241.8 \text{ kHz}$). As can be seen in Figure 7-5, because of the slightly multimode nature of the field, now there is a more complex spatial distribution with

both the amplitude and phase increasing and decreasing relative to the background level as the probe passes over the slightly multimode fiber output. This clearly demonstrates the complementary nature of NSOM and OIF collection.

Figure 7-5: This figure is for the same setup as Figure 7-4 except that the 1310 *nm* laser diode was replaced with a 635 *nm* laser diode such that the fiber is now slightly multimode. The resulting topography is shown in (a), the NSOM signal in (b) and the amplitude and phase of the signal from the tuning fork demodulated at the optical modulation frequency are shown in (c) and (d) respectively.

We point out a couple of key facts about the results presented in Figure 7-4 and Figure 7-5. The first is that we were able to use the same probe to measure optical forces at two very different wavelengths even though the photon detector used to record the NSOM signals needed to be switched between the two cases. In fact, the same probe may be used to detect an even

wider range of wavelengths. In comparison, an aperture NSOM (a-NSOM) is inherently narrowband, limited by the low transmission through the aperture for long wavelengths and a relatively low resolution relative to the wavelength at small wavelengths. Scattering NSOM (s-NSOM) does allow for broadband scattering of the signal with a single probe but the scattered radiation must then be detected using by a wavelength selective detector. Thus, in contrast to standard NSOM techniques, the measurement modality introduced in this chapter allows for broadband *sensing* with a single detector as well as sensing at wavelengths for which may be impractical to operate other types of photodetectors, such as the far IR and THz [74,75].

Second, we note that the strength of the signal is much stronger when the tip of the probe is directly over the regions of higher light intensity than after it passes over these areas. This indicates that the response is dominated by the fields near the tip of the probe rather than light coupled farther up the shaft. The result is particularly noteworthy because largely propagating light was imaged in these experiments we used primarily propagating optical fields. Consequently, MF-NSOM measurements provide a localized and thus high spatial resolution measurement of the optical signal, even in the presence of significant propagating fields. This localization is indeed much stronger with Cr coated AFM probes (not shown) where the in-contact signal is much stronger and the out-of-contact signal is negligible, as we discuss later.

7.4 MF-NSOM Imaging

The multi-frequency NSOM based on measuring optically induced forces can provide images with sub-wavelength resolution. To demonstrate this capability, we placed a gold nanosphere lithography (NSL) sample over the face of a single mode fiber illuminated at 1550 nm , as shown in Figure 7-6(a). The average intensity incident on the NSL sample at the

center of the illumination spot was $0.28 \text{ mW}/\mu\text{m}^2$. The NSL sample was made using $0.453 \mu\text{m}$ diameter spheres which corresponds to triangle sizes of $\sim 105 \text{ nm}$ having a center to center spacing of $\sim 260 \text{ nm}$ [123]. Figure 7-6(b) shows the topography of the nanosphere lithography sample. The incident light was modulated at 218.43 kHz , which is near the higher order resonance of the Cr coated AFM probe used in this scan. In Figure 7-6(c) and (d) we show the amplitude and phase measured at the optical driving frequency. To better clarify the location of the signals relative to the topographic features of the sample, we present in Figure 7-6(e) and (f) the topography as a 3-D relief and then superimpose the amplitude and phase data as the color map. As can be seen, the measured amplitude is lowest over the Au pads, moderate over the glass and it is highest near the sharp edges of the NSL sample. The phase, on the other hand, follows the sample topography. The differences in the phase signal are due to the force gradients shifting the resonance frequency. The presence of a higher oscillation amplitude over the glass areas is simply due to the higher relative optical intensities compared to the regions covered by the Au pads.

Figure 7-6: Experimental schematic (a) and the measured NSL topography (b) as described in text. Measured amplitude (c) and phase (d) of the tuning fork signal at the optical driving frequency. Color coded amplitude (e) and phase (f) mapped on the 3D topography of NSL.

The large amplitude signals near the edges can have three possible origins. First, the optically induced forces are highest and thus drive the tuning fork the strongest. Second, the tip-

sample separation changes momentarily near the edges as we scan across the surface which may result in an imaging artifact. However, in this case we would expect the signal to be highest when the probe goes from glass to gold and lowest when the probe goes from gold to glass; thus, the forward and backward (not shown) scans should show opposite locations of peak amplitude. In fact, they were the same in both forward and backward scans ruling out the possibility of such an artifact. Another possibility is an increased force gradient near the edges of the pads can cause the tuning fork resonance frequency to shift thus changing the effective spring constant and/or damping at the excitation frequency. Because the tuning fork is excited very close to resonance, a shift in the resonance frequency can only cause the effective spring constant at the excitation frequency to decrease; similarly the damping can only increase as the probe interacts with the sample surface likewise causing the effective spring constant to decrease. Therefore, gradient induced force effects are only expected to reduce the probe's sensitivity. Also if such force gradient induced effects were present, we should expect to see a change in phase at the locations of the hot spots, which we do not. Because the signal increases near these hot spots, we may therefore conclude that the detected amplitude signal is a measure of the local intensity distribution.

The results shown in Figure 7-6 were obtained using a Cr coated AFM probe rather than an uncoated NSOM probe. This allows us to achieve higher resolution as compared to an NSOM probe (diameter 20 *nm* vs 100 *nm*). Second, the signal from the Cr coated AFM probes is both significantly stronger than the uncoated probes when the probe is in contact with the sample surface and much lower out of contact. This difference originates from the smaller scattering cross section of the Cr coated AFM probe which minimizes the influence of radiation pressure

due to propagating waves when the probe is out of contact. There are two possible explanations for this effect. First, near the surface, the Cr coated probe confers stronger optical binding due to its higher refractive index contrast. This would mean that the volume of interaction is quite small for Cr coated AFM probes even in the presence of propagating radiation; thus, high resolution may be obtained. A second possibility could be the influence of an electrostatic force between the metal coated probe and the surface, as it occurs in Kelvin Probe Force Microscopy [54,56,119].

7.5 Discussion

Naturally when using metal coated probes, one must question the influence of thermal effects in the measurement. There are a few possible mechanisms through which thermal effects can manifest themselves. First, thermal effects (thermal tip elongation, photophoresis etc.) can occur at the frequency of interest. We can safely rule out this possibility since we modulate the light at $\sim 200\text{ kHz}$, which is well above the thermal response times which are typically of the order of milliseconds [27,71]. Second, there may be a possibility that thermally induced effects present at a lower frequency might couple into the higher order resonances we examine. The two additional major frequencies present are the electrical driving frequency used for feedback control of $30-40\text{ kHz}$, which is still too high for thermal effects to follow, and the zero frequency component where effects such as probe elongation can occur. Certainly, small changes in the shape of the probe may affect its mechanical behavior but such an effect is expected to be minimal in the frequency range of our measurements. This is in contrast to the electrical modulation channel where such thermal effects may influence the measured result since feedback is applied to the electrical modulation channel.

We emphasize that the measured results are not specific to our tuning fork based NSOM system but are rather a general phenomenon, observable for both tuning fork and cantilever based systems. To demonstrate the general nature of our results, one of our colleagues repeated the experiment using a cantilever based system (WITec alpha300 S) [124]. The only differences in comparison with the previous measurement were the operation wavelength which was 633 nm and the optical modulation frequency which was 109 Hz , which is significantly below the lowest mechanical resonance of the probe. We observed quite similar amplitude distributions, specifically the amplitude was lowest over the pads, moderate over glass and highest near the edges [125].

The question logically arises as to how this new measurement modality, MF-NSOM, compares to a-NSOM and s-NSOM in terms of sensitivity and performance. A direct comparison is difficult because sensing in these two modalities arises from very different physical mechanisms, specifically energy detection in the case of standard NSOM and the mechanical action of optical fields in the case of MF-NSOM. In some respects, MF-NSOM is similar to s-NSOM and thus shares many of its advantages over a-NSOM, including the fact that the achievable spatial resolution is limited by the size of the probe rather than by the fraction of a wavelength necessary to get sufficient light transmission through the probe's aperture. Thus both MF-SNOM and s-NSOM have a broader range of wavelengths with which a single probe will interact. It should be emphasized though that while the tip of s-NSOM probe may scatter a broad range of wavelengths, the performance is still limited by the ability to detect that optical radiation, this time in the far-field of the sample. As demonstrated here, the MF-NSOM approach permits true broadband detection with a single measurement system; this detection may even be

extended to wavelengths for which other sensors may be impractical to use, such as the far IR and THz.

The use of metallic probes in near-field measurements is a notorious complication due to associated thermal effects. In this context, an additional benefit of MF-NSOM is the possibility to collect near-field data with either metal coated or uncoated probes. Combined with the high modulation frequency used to detect the optical force, this means that thermal effects may be expected to be negligibly small in the optical force detection channel.

7.6 Conclusions

We have demonstrated a novel measurement technique that allows detecting near-field optical radiation with high spatial resolution by exciting the probe's oscillation at two different frequencies: one driven electrically and providing the typical AFM feedback and another one provided directly by optically-induced forces acting on the probe. We have also shown that such multi-frequency near-field scanning measurements (MF-NSOM) can be performed with both tuning fork and cantilever based systems. This operation modality provides an attractive alternative to standard NSOM in cases where common detection of optical radiation is practically cumbersome, such as in the long wavelength regime, or when it is desirable to use a single probe for sensing a broad range of wavelengths, including those in the far IR and THz.

8 SUMMARY OF ORIGINAL CONTRIBUTIONS AND CONCLUSIONS

The pursuit of designing devices which are faster, smaller and cheaper is increasingly driving research and innovation, leading to an explosion of work in the field of nanotechnology, including nanobiotechnology. Critical to this pursuit are tools to characterize these nanostructured devices. Light microscopy enables low cost, high throughput, nondestructive sample imaging. However the resolution is limited to the diffraction limit of light, or about 250 nm . One promising technique to improve this resolution is Near-Field Scanning Optical Microscopy (NSOM), which relies on the interaction of a sharp probe with high spatial resolution evanescent near-fields located in the vicinity of the sample surface.

This interaction has enabled sub-diffraction limited imaging, but proper interpretation and quantification of the collected images demands a thorough understanding of the complex tip-sample-light interaction. For instance, previous studies have demonstrated z-motion and thermally induced effects present in the interaction which may influence or even overwhelm the desired NSOM signal.

In order to collect high resolution images, a feedback loop is typically employed to ideally maintain a constant tip-sample separation close to the surface. Because the light detection relies on counting photons whereas the feedback relies on maintaining a constant force gradient on the probe, these two channels are often assumed to be independent. However, light in addition to consisting of photons which may be counted by a photodetector also exerts a force on the probe with which it interacts. We have shown that at moderate light intensities commonly employed in NSOM, the strength of light induced force is comparable to that of typical surface

forces¹. Thus, the presence of these forces introduces an artificial “topography of light” superimposed on the actual surface topography. Consequently the tip-sample separation is modified, meaning that the light is measured at a different location in the exponentially decaying near fields. Furthermore the strength of this effect depends on the local strength and gradient of the electromagnetic fields. This means the tip may follow the sample surface globally with only local deviations where the fields are the strongest – typically exactly the locations of peak interest. Thus care should be exercised to ensure the feedback is dominated by the surface forces at all location in the scan or at a minimum that all deviations are well characterized.

While the sensitivity of the probe to the local optically induced force gradients may be undesirable in typical NSOM experiments, force detection provides unique opportunities for device characterization. For example, the strength of the optically induced force gradient may be quantitatively determined for either propagating or evanescent fields by recording topography maps at several illumination intensities². Notably, during this procedure, one can also determine the tip-sample separation at closest approach, a quantity which is otherwise cumbersome to obtain. Not only does this permit the quantitative determination of the strength of optically induced force gradient acting on the probe, but a well characterized light distribution may likewise be used to characterize other unknown forces such as, for instance, the Casimir force in a nanostructured optomechanical devices.

¹ Dana C. Kohlgraf-Owens, Sergey Sukhov, and Aristide Dogariu, “Optical-force-induced artifacts in scanning probe microscopy”, *Opt. Lett.* 36, 4758-4760 (2011).

² Dana C. Kohlgraf-Owens, Sergey Sukhov, and Aristide Dogariu, “Mapping the mechanical action of light”, *Phys. Rev. A*, 84, 011807(R) (2011).

Controlled studies of the interaction between the force gradient and the scanning probe demonstrate that the specific field components measured which interact with the probe are complementary to the components collected with the probe aperture in a typically NSOM experiment³. Thus, two-dimensional scans (images) obtained from the force gradient and NSOM detection may be combined in a single measurement to provide two complementary measures of the 3D field distribution. This permits the collection of both transverse and longitudinal fields in a single measurement.

Finally the scanning probe may be used to measure not only the gradient of the optically induced forces but also the force itself⁴. This is accomplished by modulating the optical field to drive the oscillation of the probe at a frequency near a tuning fork resonance. In this manner, the light is detected not in the far-field by a photon counting detector but rather locally by the piezoelectrically generated current the force induces on the tuning fork. This permits the simultaneous acquisition of the topography and optical force at two separate frequencies. In addition, because a tuning fork has a high Q, it effectively filters radiation which is excited off resonance. Thus, the tuning fork acts as an inexpensive, compact lock-in detector.

In addition to the unique opportunities offered by the detection of light induced forces and force gradients in the visible domain, it should also be noted the additional benefit of detection over a very broad range of wavelengths, including the THz and infrared. This type of

³ Dana C. Kohlgraf-Owens, Sergey Sukhov, and Aristide Dogariu, "Discrimination of Field Components in Optical Probe Microscopy", *Opt. Lett.*, 37, 3606 (2012).

⁴ D. Kohlgraf-Owens, L. Greusard, S. Sukhov, Y. De Wilde, and A. Dogariu, "Multi-Frequency Near-Field Scanning Optical Microscopy," submitted, (2013).

local field detection is of particular interest in this long wavelength regime where the range of available detectors is limited.

In closing, by careful and systematic NSOM measurements, we demonstrated the presence and the utility of optically induced forces that on scanning probes. Most of the findings are rather general and can be applied to other scanning modalities. Optimized design of probe geometry and experimental configurations is expected to lead to increased force sensitivity, making, particularly the direct force detection technique introduced in Chapter 7, a viable alternative to standard NSOM imaging modalities.

APPENDIX: LIST OF PUBLICATIONS AND PRESENTATIONS

8.1 Refereed Publications

- [8] D. Kohlgraf-Owens, L. Greusard, S. Sukhov, Y. De Wilde, and A. Dogariu, "Multi-Frequency Near-Field Scanning Optical Microscopy," submitted, (2013).
- [7] D. C. Kohlgraf-Owens, S. Sukhov, and A. Dogariu, "Near-Field Topography of Light", Optics and Photonics News 23, 39 (2012)
- [6] Dana C. Kohlgraf-Owens, Sergey Sukhov, and Aristide Dogariu, "Discrimination of Field Components in Optical Probe Microscopy", Opt. Lett, 37, 3606 (2012).
Selected to appear in Vol. 7, Iss. 11 Virtual Journal for Biomedical Optics
- [5] Dana C. Kohlgraf-Owens, Sergey Sukhov, and Aristide Dogariu, "Optical-force-induced artifacts in scanning probe microscopy", Opt. Lett. 36, 4758-4760 (2011).
Selected to appear in: Vol. 7, Iss. 2 Virtual Journal for Biomedical Optics
- [4] Dana C. Kohlgraf-Owens, Sergey Sukhov, and Aristide Dogariu, "Mapping the mechanical action of light", Phys. Rev. A, 84, 011807(R) (2011).
- [3] Dana C. Kohlgraf-Owens, David Haefner, Sergey Sukhov, and Aristide Dogariu, "Intrinsic detection of scattering phase with near-field scanning optical microscope", Opt. Lett. 35, 2463-2465 (2010).
- [2] Dana C. Kohlgraf-Owens and Pieter G. Kik, "Structural control of nonlinear optical absorption and refraction in dense metal nanoparticle arrays", Opt. Express 17, 15032-15042 (2009).
- [1] Dana C. Kohlgraf-Owens and Pieter G. Kik, "Numerical study of surface plasmon enhanced nonlinear absorption and refraction", Opt. Express 16, 10823-10834 (2008).

8.2 Contributed Presentations

- [11] D. C. Kohlgraf-Owens, L. Greusard, S. Sukhov, Y. De Wilde, and A. Dogariu, “Broadband Near-Field Detection with Multifrequency Probe Microscopy”, *Quantum Electronics and Laser Science Conference*, (submitted).
- [10] D. C. Kohlgraf-Owens, L. Greusard, S. Sukhov, R. Colombelli, Y. De Wilde, and A. Dogariu, "Optical Multifrequency Scanning Probe Microscopy," in *Frontiers in Optics Conference*, OSA Technical Digest (Optical Society of America, 2012), paper FW2F.3.
- [9] D. C. Kohlgraf-Owens, L. Greusard, S. Sukhov, R. Colombelli, Y. De Wilde, and A. Dogariu, “The topography of light”, accepted The Twelfth International Conferences on Near-Field Optics, Nanophotonics and Related Techniques, 2012.
- [8] D. C. Kohlgraf-Owens, S. Sukhov, and A. Dogariu, “Optical Force Induced Artifacts in NSOM Measurements”, in *Frontiers in Optics*, OSA Technical Digest (Optical Society of America, 2011), paper FWQ6.
- [7] D. C. Kohlgraf-Owens, S. Sukhov, and A. Dogariu, “Exploiting Optical Forces to Characterize Electromagnetic Fields”, in *Frontiers in Optics*, OSA Technical Digest (Optical Society of America, 2011), paper FTuL4.
- [6] D. Kohlgraf-Owens, S. Sukhov, and A. Dogariu, “Optical Forces in Scanning Probe Microscopy”, in *Quantum Electronics and Laser Science Conference*, OSA Technical Digest (CD) (Optical Society of America, 2011), paper QThL4.
- [5] D. C. Kohlgraf-Owens, D. Haefner, S. Sukhov, and A. Dogariu, “Sub-Surface Interferometric Near-Field Tomography”, in *Computational Optical Sensing and Imaging*, OSA Technical Digest (CD) (Optical Society of America, 2009), paper CThC1.

- [4] D. C. Kohlgraf-Owens and P. G. Kik, “Linear and Nonlinear Effective Medium Properties of Metallodielectric Composites of Interacting Spheres and Isolated Spheroids”, in *Plasmonics and Metamaterials*, OSA Technical Digest (CD) (Optical Society of America, 2008), paper MThB3.
- [3] D. C. Kohlgraf-Owens and P. G. Kik, “Linear and Nonlinear Optical Properties of Nanostructured Metallodielectric Composites”, NanoFlorida, 2008.
- [2] Kohlgraf-Owens, D.C.; Kik, P. G.; “Surface Plasmon Mediated Optical Limiting in Dilute Metallodielectric Systems”, *Lasers and Electro-Optics Society, 2007. LEOS 2007. The 20th Annual Meeting of the IEEE*, pp.753-754, 21-25 Oct. 2007.
- [1] Walton, E.; Lee, E.; Kohlgraf, D.; Pavlovicz, R.; Bruce, G.; Montgomery, B.; , “Compact Shipboard Antenna System for Simultaneous Communication with Three Separate Satellites”, *OCEANS, 2005. Proceedings of MTS/IEEE*, pp.1577-1580 Vol. 2, 17-23 Sept. 2005.

8.3 Contributed Posters

- [2] L. Greusard, D. C. Kohlgraf-Owens, S. Sukhov, R. Colombelli, Y. De Wilde, and A. Dogariu, “Optical multifrequency atomic force microscopy”, accepted The Twelfth International Conferences on Near-Field Optics, Nanophotonics and Related Techniques, 2012.
- [1] L. A. Padilha, J. Fontana, D. Kohlgraf-Owens, M. Moreira, S. Webster, P. Pálffy-Muhoray, P. G. Kik, D. J. Hagan, and E. W. Van Stryland, “Linear and Nonlinear Optical Response of Aligned Gold Nanorods”, in *Conference on Lasers and Electro-Optics/International Quantum Electronics Conference*, OSA Technical Digest (CD) (Optical Society of America, 2009), paper JTuD123.

REFERENCES

1. P. R. Norton, "Photodetectors," in *Handbook of Optics* (McGraw Hill, 2010), Vol. 2.
2. Bahaa E. A. Saleh and M. C. Teich, *Fundamentals of Photonics*, 2nd ed. (John Wiley & Sons, 2007).
3. M. Fernández-Suárez and A. Y. Ting, "Fluorescent probes for super-resolution imaging in living cells," *Nature Reviews Molecular Cell Biology* **9**, 929–943 (2008).
4. B. Huang, W. Wang, M. Bates, and X. Zhuang, "Three-Dimensional Super-Resolution Imaging by Stochastic Optical Reconstruction Microscopy," *Science* **319**, 810–813 (2008).
5. S. W. Hell, "Far-Field Optical Nanoscopy," *Science* **316**, 1153–1158 (2007).
6. A. Rogalski and F. Sizov, "Terahertz detectors and focal plane arrays," *Opto-Electronics Review* **19**, 346–404 (2011).
7. W. L. Wolfe and P. W. Kruse, "Thermal Detectors," in *Handbook of Optics* (McGraw Hill, 2010), Vol. 2.
8. S. T. Huntington, P. G. Hartley, and J. Katsifolis, "Application of evanescent wave optics to the determination of absolute distance in surface force measurements using the atomic force microscope," *Ultramicroscopy* **94**, 283–291 (2003).
9. S. C. Clark, J. Y. Walz, and W. A. Ducker, "Atomic Force Microscopy Colloid–Probe Measurements with Explicit Measurement of Particle–Solid Separation," *Langmuir* **20**, 7616–7622 (2004).
10. U. C. Fischer, "Scanning Near-Field Optical Microscopy," in *Scanning Probe Microscopy: Analytical Methods* (Springer, n.d.), pp. 161–210.
11. M. Ohtsu and H. Hori, *Near-Field Nano-Optics: From Basic Principles to Nano-Fabrication and Nano-Photonics*, 1st ed. (Kluwer Academic/Plenum Publishers, 1999).
12. Nanonics Company, "Fiber Probes," <http://nanonics.co.il/spm-probes-and-nanotools.html>.
13. D. Cox, "The Snowman," <http://info.ee.surrey.ac.uk/Personal/D.Cox/page7/page7.html>.
14. L. Novotny and B. Hecht, *Principles of Nano-Optics* (Cambridge University Press, 2006).
15. B. Cappella and G. Dietler, "Force-distance curves by atomic force microscopy," *Surface Science Reports* **34**, 1–104 (1999).

16. H.-J. Butt, B. Cappella, and M. Kappl, "Force measurements with the atomic force microscope: Technique, interpretation and applications," *Surface Science Reports* **59**, 1–152 (2005).
17. T. D. Stowe, K. Yasumura, T. W. Kenny, D. Botkin, K. Wago, and D. Rugar, "Attonewton force detection using ultrathin silicon cantilevers," *Applied Physics Letters* **71**, 288–290 (1997).
18. F. J. Giessibl, "Advances in atomic force microscopy," *Rev. Mod. Phys.* **75**, 949–983 (2003).
19. C. Grimellec, P.-E. Milhiet, E. Perez, F. Pincet, J.-P. Aimé, V. Emiliani, O. Thoumine, T. Lionnet, V. Croquette, J.-F. Allemand, and D. Bensimon, "Nanoforce and Imaging," in *Nanoscience*, P. Boisseau, P. Houdy, and M. Lahmani, eds. (Springer Berlin Heidelberg, 2009), pp. 375–475.
20. Y. Seo and W. Jhe, "Atomic force microscopy and spectroscopy," *Reports on Progress in Physics* **71**, 016101 (2008).
21. T. R. Albrecht, P. Grütter, D. Horne, and D. Rugar, "Frequency modulation detection using high - Q cantilevers for enhanced force microscope sensitivity," *Journal of Applied Physics* **69**, 668–673 (1991).
22. B. Gotsmann, C. Seidel, B. Anczykowski, and H. Fuchs, "Conservative and dissipative tip-sample interaction forces probed with dynamic AFM," *Phys. Rev. B* **60**, 11051–11061 (1999).
23. N. Satoh, T. Fukuma, K. Kobayashi, S. Watanabe, T. Fujii, K. Matsushige, and H. Yamada, "Near-field light detection by conservative and dissipative force modulation methods using a piezoelectric cantilever," *Applied Physics Letters* **96**, 233104–233104–3 (2010).
24. S. K. Buratto, J. W. P. Hsu, J. K. Trautman, E. Betzig, R. B. Bylisma, C. C. Bahr, and M. J. Cardillo, "Imaging InGaAsP quantum-well lasers using near-field scanning optical microscopy," *Journal of Applied Physics* **76**, 7720–7725 (1994).
25. C. Lienau, A. Richter, and T. Elsaesser, "Light - induced expansion of fiber tips in near - field scanning optical microscopy," *Applied Physics Letters* **69**, 325–327 (1996).
26. A. Ambrosio, O. Fenwick, F. Cacialli, R. Micheletto, Y. Kawakami, P. G. Gucciardi, D. J. Kang, and M. Allegrini, "Shape dependent thermal effects in apertured fiber probes for scanning near-field optical microscopy," *Journal of Applied Physics* **99**, 084303–084303–6 (2006).

27. P. G. Gucciardi, M. Colocci, M. Labardi, and M. Allegrini, "Thermal-expansion effects in near-field optical microscopy fiber probes induced by laser light absorption," *Applied Physics Letters* **75**, 3408–3410 (1999).
28. M. Nonnenmacher and H. K. Wickramasinghe, "Optical absorption spectroscopy by scanning force microscopy," *Ultramicroscopy* **42-44, Part 1**, 351–354 (1992).
29. B. Hecht, B. Sick, U. P. Wild, V. Deckert, R. Zenobi, O. J. F. Martin, and D. W. Pohl, "Scanning near-field optical microscopy with aperture probes: Fundamentals and applications," *The Journal of Chemical Physics* **112**, 7761–7774 (2000).
30. C. Obermüller and K. Karrai, "Far field characterization of diffracting circular apertures," *Applied Physics Letters* **67**, 3408–3410 (1995).
31. H. A. Bethe, "Theory of Diffraction by Small Holes," *Phys. Rev.* **66**, 163–182 (1944).
32. C. J. Bouwkamp, "Diffraction Theory," *Reports on Progress in Physics* **17**, 35–100 (1954).
33. T. David, C. Chicanne, N. Richard, J. R. Krenn, F. Scheurer, K. Ounadjela, M. Hehn, Y. Lacroute, and J. P. Goudonnet, "Application to dielectric, metallic, and magnetic samples of a transmission mode scanning near field optical microscope with normal force distance regulation on bent optical fibers," *Review of Scientific Instruments* **70**, 4587–4594 (1999).
34. D. C. Kohlgraf-Owens, D. Haefner, S. Sukhov, and A. Dogariu, "Intrinsic detection of scattering phase with near-field scanning optical microscope," *Opt. Lett.* **35**, 2463–2465 (2010).
35. K. D. Weston and S. K. Buratto, "A Reflection Near-Field Scanning Optical Microscope Technique for Subwavelength Resolution Imaging of Thin Organic Films," *J. Phys. Chem. B* **101**, 5684–5691 (1997).
36. F. Keilmann and R. Hillenbrand, "Near-Field Microscopy by Elastic Light Scattering from a Tip," *Philosophical Transactions: Mathematical, Physical and Engineering Sciences* **362**, 787–805 (2004).
37. B. Hecht, H. Bielefeldt, Y. Inouye, D. W. Pohl, and L. Novotny, "Facts and artifacts in near-field optical microscopy," *Journal of Applied Physics* **81**, 2492–2498 (1997).
38. J.-J. Greffet and R. Carminati, "Image formation in near-field optics," *Progress in Surface Science* **56**, 133–237 (1997).
39. R. Carminati, A. Madrazo, M. Nieto-Vesperinas, and J.-J. Greffet, "Optical content and resolution of near-field optical images: Influence of the operating mode," *Journal of Applied Physics* **82**, 501–509 (1997).

40. I. P. Radko, V. S. Volkov, S. I. Bozhevolnyi, J. Henningsen, and J. Pedersen, "Near - field mapping of surface refractive - index distributions," *Laser Physics Letters* **2**, 440–444 (2005).
41. M. Labardi, S. Patanè, and M. Allegrini, "Artifact-free near-field optical imaging by apertureless microscopy," *Applied Physics Letters* **77**, 621–623 (2000).
42. C. E. Jordan, S. J. Stranick, L. J. Richter, and R. R. Cavanagh, "Removing optical artifacts in near-field scanning optical microscopy by using a three-dimensional scanning mode," *Journal of Applied Physics* **86**, 2785–2789 (1999).
43. R. Hillenbrand, B. Knoll, and F. Keilmann, "Pure optical contrast in scattering-type scanning near-field microscopy," *Journal of Microscopy* **202**, 77 (2001).
44. J. N. Walford, J. A. Porto, R. Carminati, J.-J. Greffet, P. M. Adam, S. Hudlet, J.-L. Bijeon, A. Stashkevich, and P. Royer, "Influence of tip modulation on image formation in scanning near-field optical microscopy," *Journal of Applied Physics* **89**, 5159–5169 (2001).
45. A. S. Foster and A. H. Werner, *Scanning Probe Microscopy: Atomic Scale Engineering by Forces and Currents*, Nanoscience and Technology (Springer, 2006).
46. D. Tabor and R. H. S. Winterton, "The Direct Measurement of Normal and Retarded van der Waals Forces," *Proceedings of the Royal Society of London. A. Mathematical and Physical Sciences* **312**, 435 –450 (1969).
47. G. L. Klimchitskaya, U. Mohideen, and V. M. Mostepanenko, "The Casimir force between real materials: Experiment and theory," *Rev. Mod. Phys.* **81**, 1827–1885 (2009).
48. H.-J. Butt and M. Kappl, *Surface and Interfacial Forces* (2010).
49. A. W. Rodriguez, F. Capasso, and S. G. Johnson, "The Casimir effect in microstructured geometries," *Nat Photon* **5**, 211–221 (2011).
50. D. A. Bonnell and R. Shao, "Principles of Basic and Advanced Scanning Probe Microscopy," in *Scanning Probe Microscopy: Characterization, Nanofabrication, and Device Application of Functional Materials*, NATO Science Series No. II: Mathematics, Physics and Chemistry (Kluwer Academic Publishers, 2005), Vol. 186, pp. 77–102.
51. E. Meyer, H. J. Hug, and R. Bennewitz, *Scanning Probe Microscopy: The Lab on a Tip* (Springer, 2004).
52. M. Nonnenmacher, M. P. OBoyle, and H. K. Wickramasinghe, "Kelvin probe force microscopy," *Applied Physics Letters* **58**, 2921–2923 (1991).

53. J. M. R. Weaver, "High resolution atomic force microscopy potentiometry," *Journal of Vacuum Science & Technology B: Microelectronics and Nanometer Structures* **9**, 1559 (1991).
54. J. Mertz, M. Hipp, J. Mlynek, and O. Marti, "Optical near - field imaging with a semiconductor probe tip," *Applied Physics Letters* **64**, 2338–2340 (1994).
55. M. Abe, Y. Sugawara, K. Sawada, Y. Andoh, and S. Morita, "Near-field optical imaging using force detection with new tip-electrode geometry," *Applied Surface Science* **140**, 383–387 (1999).
56. M. Abe, Y. Sugawara, Y. Hara, K. Sawada, and S. Morita, "Force Imaging of Optical Near-Field Using Noncontact Mode Atomic Force Microscopy," *Japanese Journal of Applied Physics* **37**, L167–L169 (1998).
57. M. Abe, "Detection mechanism of an optical evanescent field using a noncontact mode atomic force microscope with a frequency modulation detection method," *Journal of Vacuum Science & Technology B: Microelectronics and Nanometer Structures* **15**, 1512 (1997).
58. K. Ozasa, S. Nemoto, M. Maeda, and M. Hara, "Kelvin probe force microscope with near-field photoexcitation," *Journal of Applied Physics* **107**, 103501–103501–4 (2010).
59. W. Denk and D. W. Pohl, "Local electrical dissipation imaged by scanning force microscopy," *Applied Physics Letters* **59**, 2171–2173 (1991).
60. D. W. Pohl, "Photons and Forces I: Light Generates Force," in *Forces in Scanning Probe Methods*, NATO ASI Series No. E: Applied Sciences (1995), Vol. 286, pp. 235–248.
61. F. Depasse and D. Courjon, "Inductive forces generated by evanescent light fields: application to local probe microscopy," *Optics Communications* **87**, 79–83 (1992).
62. C. Girard, A. Dereux, and O. J. F. Martin, "Theoretical analysis of light-inductive forces in scanning probe microscopy," *Phys. Rev. B* **49**, 13872–13881 (1994).
63. C. Girard, "Theoretical analysis of scanning near - field optical microscopy," *Scanning* **16**, 333–342 (1994).
64. T. Iida and H. Ishihara, "Theoretical study of resonant-light-induced force microscopy," *Nanotechnology* **18**, 084018 (2007).
65. T. Iida and H. Ishihara, "Theory of light - induced force microscopy to observe collective excited states in quantum - dot - array," *physica status solidi (c)* **6**, 898–901 (2009).

66. T. Iida and H. Ishihara, "Force Control between Quantum Dots by Light in Polaritonic Molecule States," *Phys. Rev. Lett.* **97**, 117402 (2006).
67. X. Zhu, G.-S. Huang, H.-T. Zhou, X. Yang, Z. Wang, Y. Ling, Y.-D. Dai, and Z.-Z. Gan, "Ultrasonic resonance regulated near-field scanning optical microscope and laser induced near-field optical-force interaction," *Optical Review* **4**, A236–A239 (1997).
68. X. Zhu, Y. Ling, G. Huang, H. Zhou, Y. Dai, K. Wu, and Z. Gan, "Laser Induced Light-Force Interaction in the Optical Near-Field Region," *Chinese Physics Letters* **15**, 165–167 (1998).
69. J. Kohoutek, D. Dey, A. Bonakdar, R. Gelfand, A. Sklar, O. G. Memis, and H. Mohseni, "Opto-Mechanical Force Mapping of Deep Subwavelength Plasmonic Modes," *Nano Lett.* **11**, 3378–3382 (2011).
70. D. I. Kavalidjiev, R. Toledo - Crow, and M. Vaez - Iravani, "On the heating of the fiber tip in a near - field scanning optical microscope," *Applied Physics Letters* **67**, 2771–2773 (1995).
71. A. H. La Rosa, B. I. Yakobson, and H. D. Hallen, "Origins and effects of thermal processes on near - field optical probes," *Applied Physics Letters* **67**, 2597–2599 (1995).
72. M. Stähelin, M. A. Bopp, G. Tarrach, A. J. Meixner, and I. Zschokke - Gränacher, "Temperature profile of fiber tips used in scanning near - field optical microscopy," *Applied Physics Letters* **68**, 2603–2605 (1996).
73. E. Bourillot, T. David, Y. Lacroute, and E. Lesniewska, "Transversal mode and thermal analysis of an InP laser diode by near-field scanning probe microscopies," *J. Opt. Soc. Am. B* **25**, 1888–1894 (2008).
74. A. Pohlkötter, U. Willer, C. Bauer, and W. Schade, "Resonant tuning fork detector for electromagnetic radiation," *Appl. Opt.* **48**, B119–B125 (2009).
75. U. Willer, A. Pohlkötter, W. Schade, J. Xu, T. Losco, R. P. Green, A. Tredicucci, H. E. Beere, and D. A. Ritchie, "Resonant tuning fork detector for THz radiation," *Opt. Express* **17**, 14069–14074 (2009).
76. O. Marti, A. Ruf, M. Hipp, H. Bielefeldt, J. Colchero, and J. Mlynek, "Mechanical and thermal effects of laser irradiation on force microscope cantilevers," *Ultramicroscopy* **42-44, Part 1**, 345–350 (1992).
77. M. Allegrini, C. Ascoli, P. Baschieri, F. Dinelli, C. Frediani, A. Lio, and T. Mariani, "Laser thermal effects on atomic force microscope cantilevers," *Ultramicroscopy* **42-44, Part 1**, 371–378 (1992).

78. J. Tamayo, A. D. L. Humphris, and M. J. Miles, "Piconewton regime dynamic force microscopy in liquid," *Applied Physics Letters* **77**, 582–584 (2000).
79. J. Mertz, O. Marti, and J. Mlynek, "Regulation of a microcantilever response by force feedback," *Applied Physics Letters* **62**, 2344–2346 (1993).
80. T. Aoki, M. Hiroshima, K. Kitamura, M. Tokunaga, and T. Yanagida, "Non-contact scanning probe microscopy with sub-piconewton force sensitivity," *Ultramicroscopy* **70**, 45–55 (1997).
81. L. P. Ghislain, N. A. Switz, and W. W. Webb, "Measurement of small forces using an optical trap," *Review of Scientific Instruments* **65**, 2762–2768 (1994).
82. L. P. Ghislain and W. W. Webb, "Scanning-force microscope based on an optical trap," *Opt. Lett.* **18**, 1678–1680 (1993).
83. E.-L. Florin, A. Pralle, J. K. Heinrich Hörber, and E. H. K. Stelzer, "Photonic Force Microscope Based on Optical Tweezers and Two-Photon Excitation for Biological Applications," *Journal of Structural Biology* **119**, 202–211 (1997).
84. L. Malmqvist and H. M. Hertz, "Trapped particle optical microscopy," *Optics Communications* **94**, 19–24 (1992).
85. S. Kawata, Y. Inouye, and T. Sugiura, "Near-Field Scanning Optical Microscope with a Laser Trapped Probe," *Japanese Journal of Applied Physics* **33**, L1725–L1727 (1994).
86. V. K. Valev, A. V. Silhanek, Y. Jeyaram, D. Denkova, B. De Clercq, V. Petkov, X. Zheng, V. Volskiy, W. Gillijns, G. A. E. Vandenbosch, O. A. Aktsipetrov, M. Ameloot, V. V. Moshchalkov, and T. Verbiest, "Hotspot Decorations Map Plasmonic Patterns with the Resolution of Scanning Probe Techniques," *Phys. Rev. Lett.* **106**, 226803 (2011).
87. P. C. Chaumet, A. Rahmani, F. de Fornel, and J.-P. Dufour, "Evanescent light scattering: The validity of the dipole approximation," *Phys. Rev. B* **58**, 2310–2315 (1998).
88. M. Nieto-Vesperinas and J. J. Saenz, "Optical forces from an evanescent wave on a magnetodielectric small particle," *Opt. Lett.* **35**, 4078–4080 (2010).
89. M. Šiler and P. Zemánek, "Parametric study of optical forces acting upon nanoparticles in a single, or a standing, evanescent wave," *Journal of Optics* **13**, 044016 (2011).
90. M. Nieto-Vesperinas and J. R. Arias-Gonzalez, "Theory of Forces Induced by Evanescent Fields," *arXiv:1102.1613* (2011).
91. L. Novotny, "Forces in Optical Near-Fields," in *Near-Field Optics and Surface Plasmon Polaritons*, P. S. Kawata, ed., Topics in Applied Physics No. 81 (Springer Berlin Heidelberg, 2001), pp. 123–141.

92. P. C. Chaumet and M. Nieto-Vesperinas, "Time-averaged total force on a dipolar sphere in an electromagnetic field," *Optics Letters* **25**, 1065–1067 (2000).
93. J. R. Arias-González and M. Nieto-Vesperinas, "Optical forces on small particles: attractive and repulsive nature and plasmon-resonance conditions," *J. Opt. Soc. Am. A* **20**, 1201–1209 (2003).
94. P. C. Chaumet and M. Nieto-Vesperinas, "Electromagnetic force on a metallic particle in the presence of a dielectric surface," *Phys. Rev. B* **62**, 11185–11191 (2000).
95. M. Nieto-Vesperinas, J. J. Sáenz, R. Gómez-Medina, and L. Chantada, "Optical forces on small magnetodielectric particles," *Opt. Express* **18**, 11428–11443 (2010).
96. J. Chen, J. Ng, Z. Lin, and C. T. Chan, "Optical pulling force," *Nature Photonics* **5**, 531–534 (2011).
97. P. J. Valle, J.-J. Greffet, and R. Carminati, "Optical contrast, topographic contrast and artifacts in illumination-mode scanning near-field optical microscopy," *Journal of Applied Physics* **86**, 648–656 (1999).
98. N. E. Dickenson, E. S. Erickson, O. L. Mooren, and R. C. Dunn, "Characterization of power induced heating and damage in fiber optic probes for near-field scanning optical microscopy," *Review of Scientific Instruments* **78**, 053712–053712–6 (2007).
99. D. C. Kohlgraf-Owens, S. Sukhov, and A. Dogariu, "Mapping the mechanical action of light," *Phys. Rev. A* **84**, 011807 (2011).
100. F. J. Giessibl and H. Bielefeldt, "Physical interpretation of frequency-modulation atomic force microscopy," *Phys. Rev. B* **61**, 9968–9971 (2000).
101. J. D. Jackson, *Classical Electrodynamics*, 3rd ed. (Wiley, 1999).
102. D. Haefner, S. Sukhov, and A. Dogariu, "Conservative and Nonconservative Torques in Optical Binding," *Phys. Rev. Lett.* **103**, 173602 (2009).
103. A. Dereux, C. Girard, O. J. F. Martin, and M. Devel, "Optical Binding in Scanning Probe Microscopy," *Europhysics Letters (EPL)* **26**, 37–42 (1994).
104. P. J. van Zwol, G. Palasantzas, and J. T. M. DeHosson, "Weak dispersive forces between glass and gold macroscopic surfaces in alcohols," *Phys. Rev. E* **79**, 041605 (2009).
105. H. W. Kihm, S. M. Koo, Q. H. Kim, K. Bao, J. E. Kihm, W. S. Bak, S. H. Eah, C. Lienau, H. Kim, P. Nordlander, N. J. Halas, N. K. Park, and D.-S. Kim, "Bethe-hole polarization analyser for the magnetic vector of light," *Nat Commun* **2**, 451 (2011).

106. M. Burrelli, D. van Oosten, T. Kampfrath, H. Schoenmaker, R. Heideman, A. Leinse, and L. Kuipers, "Probing the Magnetic Field of Light at Optical Frequencies," *Science* **326**, 550–553 (2009).
107. D. J. Griffiths, *Introduction to Electrodynamics*, 3rd ed. (Pearson Education, 1999).
108. C. Bohren and D. Huffman, *Absorption and Scattering of Light by Small Particles*, 2nd ed. (WILEY_VCH, 2012).
109. "Numerical simulations using COMSOL Multiphysics indicate that the results do not qualitatively change for larger particles or due to any plasmonic effects.," (n.d.).
110. D. C. Kohlgraf-Owens, S. Sukhov, and A. Dogariu, "Optical-force-induced artifacts in scanning probe microscopy," *Optics Letters* **36**, 4758 (2011).
111. R. Garcia and E. T. Herruzo, "The emergence of multifrequency force microscopy," *Nature Nanotechnology* **7**, 217–226 (2012).
112. R. Proksch, "Multi-Frequency Atomic Force Microscopy," in *Scanning Probe Microscopy of Functional Materials: Nanoscale Imaging and Spectroscopy* (Springer, 2010).
113. D. Forchheimer, D. Platz, E. A. Tholén, and D. B. Haviland, "Model-based extraction of material properties in multifrequency atomic force microscopy," *Phys. Rev. B* **85**, 195449 (2012).
114. D. C. Kohlgraf-Owens, S. Sukhov, and A. Dogariu, "Discrimination of field components in optical probe microscopy," *Opt. Lett.* **37**, 3606–3608 (2012).
115. R. C. Davis, C. C. Williams, and P. Neuzil, "Optical intensity mapping on the nanometer scale by near-field photodetection optical microscopy," *Opt. Lett.* **21**, 447–449 (1996).
116. H. U. Danzebrink, O. Ohlsson, and G. Wilkening, "Fabrication and characterization of optoelectronic near-field probes based on an SFM cantilever design," *Ultramicroscopy* **61**, 131–138 (1995).
117. P. Bauer, B. Hecht, and C. Rossel, "Piezoresistive cantilevers as optical sensors for scanning near-field microscopy," *Ultramicroscopy* **61**, 127–130 (1995).
118. I. Rajapaksa, K. Uenal, and H. K. Wickramasinghe, "Image force microscopy of molecular resonance: A microscope principle," *Applied Physics Letters* **97**, 073121–073121–3 (2010).
119. J. M. R. Weaver, L. M. Walpita, and H. K. Wickramasinghe, "Optical absorption microscopy and spectroscopy with nanometre resolution," *Nature* **342**, 783–785 (1989).
120. J.-M. Friedt and E. Carry, "Introduction to the quartz tuning fork," *American Journal of Physics* **75**, 415 (2007).

121. A. G. T. Ruiter, J. A. Veerman, K. O. van der Werf, and N. F. van Hulst, "Dynamic behavior of tuning fork shear-force feedback," *Applied Physics Letters* **71**, 28–30 (1997).
122. K. Karrai, "Lecture notes on shear and friction force detection with quartz tuning forks," Work presented at the "Ecole Thématique du CNRS" on near-field optics, (La Londe les Maures, France) (2000).
123. J. C. Hulteen, D. A. Treichel, M. T. Smith, M. L. Duval, T. R. Jensen, and R. P. Van Duyne, "Nanosphere Lithography: Size-Tunable Silver Nanoparticle and Surface Cluster Arrays," *J. Phys. Chem. B* **103**, 3854–3863 (1999).
124. L. Greusard, personal communication (2012).
125. D. Kohlgraf-Owens, L. Greusard, S. Sukhov, Y. De Wilde, and A. Dogariu, "Multi-Frequency Near-Field Scanning Optical Microscopy," submitted (2013).