Exploring the Nonlinear Dynamics of Tapping Mode Atomic Force Microscopy with Capillary Layer Interactions

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(ABSTRACT)

Central to tapping mode atomic force microscopy is an oscillating cantilever whose tip interacts with a sample surface. The tip-surface interactions are strongly nonlinear, rapidly changing, and hysteretic. We explore numerically a lumped-mass model that includes attractive, adhesive, and repulsive contributions as well as the interaction of the capillary fluid layers that cover both tip and sample in the ambient conditions common in experiment. To accomplish this, we have developed and used numerical techniques specifically tailored for discontinuous, nonlinear, and hysteretic dynamical systems. In particular, we use forward-time simulation with event handling and the numerical pseudo-arclength continuation of periodic solutions. We first use these numerical approaches to explore the nonlinear dynamics of the cantilever. We find the coexistence of three steady state oscillating solutions: (i) periodic with low-amplitude, (ii) periodic with high-amplitude, and (iii) high-periodic or irregular behavior. Furthermore, the branches of periodic solutions are found to end precisely where the cantilever comes into grazing contact with event surfaces in state space corresponding to the onset of capillary interactions and the onset of repulsive forces associated with surface contact. Also, the branches of periodic solutions are found to be separated by windows of irregular dynamics. These windows coexist with the periodic branches of solutions and exist beyond the termination of the periodic solution. We also explore the power dissipated through the interaction of the capillary fluid layers. The source of this dissipation is the hysteresis in the conservative capillary force interaction. We relate the power dissipation with
the fraction of oscillations that break the fluid meniscus. Using forward-time simulation with event handling, this is done exactly and we explore the dissipated power over a range of experimentally relevant conditions. It is found that the dissipated power as a function of the equilibrium cantilever-surface separation has a characteristic shape that we directly relate to the cantilever dynamics. We also find that despite the highly irregular cantilever dynamics, the fraction of oscillations breaking the meniscus behaves in a fairly simple manner. We have also performed a large number of forward-time simulations over a wide range of initial conditions to approximate the basins of attraction of steady oscillating solutions. Overall, the simulations show a complex pattern of high and low amplitude periodic solutions over the range of initial conditions explored. We find that for large equilibrium separations, the basin of attraction is dominated by the low-amplitude periodic solution and for the small equilibrium separations by the high-amplitude periodic solution.
Dedication

I dedicate this dissertation to my parents, my husband, my sister, and to the memory of my grandmother!
I would like to extend my deepest gratitude and appreciation to my advisor, Dr. Mark Paul for his support and encouragement throughout these years. I appreciate his tremendous engineering insight and guidance throughout this work. I could not have imagined having a better advisor and mentor and I would like to thank him for providing me with an opportunity to work on such exciting research.

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Nomenclature

\( x \) \hspace{1cm} \text{system state vector}
\( n \) \hspace{1cm} \text{dimension of the system}
\( t \) \hspace{1cm} \text{time}
\( f \) \hspace{1cm} \text{vector field}
\( \tau \) \hspace{1cm} \text{time constant}
\( \Phi (x, t) \) \hspace{1cm} \text{flow function}
\( h \) \hspace{1cm} \text{scalar-valued discontinuity trigger function, water film thickness}
\( i, j \) \hspace{1cm} \text{index variable}
\( i_a, i_b \) \hspace{1cm} \text{index variable after the triggered event, index variable before the triggered event}
\( P \) \hspace{1cm} \text{Poincaré map}
\( m \) \hspace{1cm} \text{mass of the cantilever}
\( k, K \) \hspace{1cm} \text{spring constant}
\( c \) \hspace{1cm} \text{damping coefficient}
\( d_0 \) \hspace{1cm} \text{tip-sample separation from the equilibrium rest position}
$F_{\text{int}}$ interaction force for simple example oscillator

$F_d$ amplitude of driving force

$\omega_d$ angular frequency of driving force

$q$ instantaneous displacement of the cantilever tip

$\dot{q}$ instantaneous velocity of the cantilever tip

$\ddot{q}$ instantaneous acceleration of the cantilever tip

$q_{\text{max}}$ maximum excursion of $q$

$Q$ quality factor

$\omega_0$ resonant frequency

$\phi$ phase

$A$ amplitude of oscillation

$I$ Identity matrix

$\mu$ $m$-dimensional vector of system control parameters

$p$ a system parameter

$\lambda$ pseudo-arclength parameter

$F_{ts}$ tip-surface force interactions

$F_v$ attractive long-range van der Waals forces

$F_c$ capillary forces

$F_r$ repulsive forces

$A_0$ amplitude of free oscillation

$d$ instantaneous tip-sample separation

$H$ Hamaker constant
\( R \) radius of curvature of the tip
\( a_0 \) intermolecular constant
\( E_t \) Young’s modulus of the tip
\( E_s \) Young’s modulus of the sample
\( E^* \) effective Young’s modulus of the tip and surface
\( \nu_t \) Poisson ratio of the tip
\( \nu_s \) Poisson ratio of the sample
\( d_{\text{on}} \) capillary force turns on
\( d_{\text{off}} \) capillary force turns off
\( V \) the meniscus volume
\( f_0 \) resonant frequency
\( \gamma_{sv} \) surface energy of the tip and sample
\( \gamma_{w} \) surface energy of water
\( P_{\text{max}} \) power dissipated by breaking the meniscus
\( E_{\text{tip}} \) energy lost in breaking the meniscus of the capillary fluid layer
\( f \) fraction of the oscillations breaking the meniscus
\( f_{H} \) fraction of the initial conditions yielding the high-amplitude periodic solution
\( f_{L} \) fraction of the initial conditions yielding the low-amplitude periodic solution
\( \delta q \) average fluctuation of the displacement about its equilibrium value
\( \delta \dot{q} \) average fluctuation of the velocity about its equilibrium value

\( k_B \) Boltzmann’s constant

\( T \) temperature
Chapter 1

Introduction

Atomic force microscopy (AFM) has been extensively used to study the surface topography of a wide variety of materials and to measure intramolecular forces [2–5]. An optical detection technique is often used to monitor the amplitude of oscillation, frequency, and phase of the AFM cantilever. During scanning, a particular operating parameter such as amplitude is maintained constant, and images are generated using a feedback loop between the optical detection system and the piezoelectric scanners. To produce topographic images of a sample, contact mode, non-contact mode, and tapping mode can be used.

In contact mode, the probe is pulled across the surface of the sample. During scanning, a constant deflection of the cantilever is maintained corresponding to a displacement of the cantilever tip relative to an undeflected cantilever. As the topography of the sample changes, the scanner moves the relative position of the tip with respect to the sample to maintain this constant deflection. Using this feedback mechanism, the topography of the sample is mapped during scanning. In order to minimize the amount of applied force, low spring
constant cantilevers are often used ($k \lesssim 1 \text{ N/m}$). However, significant deformation and damage of soft samples often occurs during contact mode imaging.

In non-contact mode operation of AFM, the cantilever is oscillated near its resonance frequency and does not make contact with the surface. Both the tip-sample separation and the oscillation amplitude are typically on the order of 1-10 nm. The tip oscillates above the surface. However, because most samples are covered by a liquid layer, keeping the cantilever tip close enough to the sample for the inter-atomic forces to become detectable while preventing the tip from sticking to the surface makes this method hard to use in ambient conditions. Non-contact mode AFM is often the method of choice for ultra high vacuum conditions.

Tapping mode AFM offers advantages in surface topography measurements when compared to contact and non-contact modes. Figure 1.1 shows an electron micrograph of two commercially available tapping mode AFMs.

![AFM Cantilever](image)

Figure 1.1: An example of a commercial silicon nitride AFM cantilever available from Veeco [1]. The approximate cantilever dimensions are a length of 100µm (for the smaller cantilever) or 200µm (for the larger cantilever) measured from the base to the tip, a width of 25µm (of an individual arm), and a thickness of 0.6µm.
In tapping mode the cantilever probe makes only intermittent contact with the sample and, as a result, this can be used to reduce sample destruction during measurement. Typically, the cantilever is driven to oscillate at its fundamental frequency. However, this is not always the case, for example it has recently been shown that driving the cantilever with two frequencies can be used to control bistability [6]. The tapping mode has been widely employed to study compliant materials such as polymers, biomaterials and semiconductors [4]. These characteristics of tapping mode AFM have created a great deal of interest in applying it to the study of biological structures. Surfaces occupy much of the space in a living organism and surface biology has been difficult to investigate in the past due to the lack of appropriate technologies. However, using tapping mode AFM many of these challenges have been overcome. For example, it is now possible to image DNA, single proteins, and living cells [7–12].

Furthermore, material properties can also be measured by taking advantage of the sensitivity of tip motion to forces and force gradients. Also, the simultaneous data acquisition of oscillation amplitude, frequency, and phase shift relative to that of the drive can be used to provide a detailed description of the sample.

### 1.1 Challenges in the Modeling of Atomic Force Microscopy

The modeling and simulation of atomic force microscopy has been conducted at varying degrees of sophistication [13]. Most common, is the use of a lumped-mass model with a model of the tip-surface force interactions that captures the dynamics of the fundamental
mode. This approach has been very successful in shedding new physical insights regarding the cantilever dynamics over a range of experimental conditions. Full three-dimensional finite element simulations have also been performed to explore the role of higher order modes including the contributions from torsional and lateral excitations [14]. Models including the higher harmonics of the fundamental mode have also been used [15].

In typical operation the AFM cantilever oscillates near its natural frequency in proximity to the sample while a feedback control system keeps the oscillation amplitude constant [16]. The interaction between the AFM tip and the sample is rapidly varying (for example due to the sudden impact of the cantilever with the surface), nonlinear (due to the interaction forces between the cantilever tip and sample), and hysteretic (due to the capillary forces caused by thin films of water covering both the tip and sample). As a result, the cantilever dynamics are difficult to model, analyze, and study. For example, for a given set of parameters there often exist multiple stable solutions [16–19]. As a result, in the presence of noise the oscillating AFM cantilever can switch between solutions. It is often the case that one solution will have a larger impact velocity which can be detrimental to the sample [20]. This makes the development of a control scheme to select the desirable solution attractive [21].

The interaction forces between the tip and sample are very complex and are described by a growing and substantial literature [16–18, 22–25]. Earlier work has shown that during the operation of a tapping mode AFM two branches of stable oscillations are found as one varies the drive frequency [16, 26, 27]. The bistable behavior is due to the attractive and repulsive tip sample interactions. Garcia and San Paulo [16] investigated the cantilever dynamics as the equilibrium separation between the probe and sample was varied. This permitted an exploration of the attractive and repulsive regimes and the transition between multiple
stable solutions. Zitzler et al. [17] explored the role of capillary forces resulting from thin layers of water on the tip and sample which is common under many operating conditions due to humidity in the surrounding air. The dynamics were found to jump from a low amplitude periodic solution to a high amplitude periodic solution when a critical amplitude of oscillation was surpassed. Dankowicz et al. [18, 28] have used a discontinuity mapping analysis to study the instabilities for a variety of models without the capillary force in detail.

The dynamic mode operation of micro and nanomechanical systems such as the AFM results in energy dissipation at the nanoscale [29–32]. The energy dissipated by tip-sample interactions has been widely used for phase contrast imaging. Investigation of the dissipation in AFM can be performed at the nanoscale or the atomic level [33, 34]. At the nanoscale level, the relationship between the dissipation process and macroscopic quantities is the matter of interest [32]. Several processes such as friction, viscoelasticity, dissipative interfacial interactions, hysteresis due to the surface adhesion, or hysteresis due to the interaction between capillary fluid layers can be the source of energy dissipation [33]. In this research, we have chosen to explore the dissipation due to the hysteretic behavior of the capillary force interactions in detail.

When the AFM tip approaches the sample, a nanoscale water bridge forms between the tip and the sample because of capillary condensation as shown schematically in Figure 1.2. Several studies [16,35–37] have been conducted to understand the physics of the nanoscale water bridge because of its importance in: the fabrication of nanodevices such as dip-pen nanolithography (DPN); the determination of the properties of samples; and the phase contrast imaging of a broad range of materials. Moreover, understanding the detailed mechanism of energy dissipation will provide more insights into studies of biological systems [38]. These
studies are often performed at ambient conditions and are associated with energy dissipation. The pressure difference across the interface of a surface results in capillary condensation. This phenomenon happens when the nanoscale cracks or pores of the surface are of the same order of magnitude as the Kelvin radius [39–41]. The water film thickness resulting from capillary condensation depends on the relative humidity. Previous work on quantifying the relation of relative humidity to the adsorbed water film has shown that the dependance is strongly nonlinear [42]. Approaching the sample, the meniscus forms at some specific distance from the sample and while retracting, it breaks at a larger distance from the sample [17]. The formation and rupture of the capillary bridge at different separation distances is the origin of the energy dissipation [17,37,42,43].

Figure 1.2: (color) A schematic of an AFM tip oscillating above a sample in ambient conditions. Here, the AFM tip is represented by a sphere in close proximity of the sample, an infinite surface. Blue regions show the water layer. When the tip and sample are in contact, water in the overlapping region is accumulated at the edge of the contact area. This volume of water forms a meniscus when the tip-sample separation is less than a specific value.

In order to understand the global behavior of a system, it is useful to know the collections of initial conditions that lead to all of the steady states. Initial states that evolve to an
attractor in state space form its basin of attraction. The basin boundary can be a fractal or smooth curve and fractal basins (also called riddled basins) are often found in dissipative systems [44]. In the classical analysis of nonlinear systems once the equation of motion is known, the possible equilibrium states and periodic motions of the system are found and the stability of the solutions is studied. It is also useful to quantify the evolution of the equilibrium states and periodic solutions as a function of system parameters. A more global understanding is given by the basins of attraction of these solutions. Knowledge of the basin structure is useful for precision measurement applications, such as nanomechanical bifurcation amplifiers and a sensitive monitoring of intrinsic device noise processes [45]. For example, Marth et al. [46] and Garcia and San Paulo [3] have used numerical calculations of the basins of attraction to explore the nonlinear dynamics of the AFM.

1.2 Objectives

Overall, the objectives of this dissertation are:

1. To develop and use numerical methods tailored for nonlinear dynamical systems with rapidly varying and discontinuous vector fields. We use two different numerical approaches. The first is forward-time simulation. In this case we numerically integrate the equations of motion forward in time using adaptive time stepping and event-handling capabilities. The second is numerical continuation. The continuation of periodic solutions offers some significant advantages over forward-time simulation that will be explored.

2. To explore the nonlinear dynamics of a tapping mode AFM with the inclusion of cap-
illary layer interactions. The major contribution to the dynamics is the interaction of the cantilever tip with the sample. As a result, the cantilever is often modeled as a lumped mass system with discontinuous, nonlinear, and hysteretic force interactions [16,17,47,48].

3. To explore discontinuity induced bifurcations. Using our numerical approaches, we demonstrate the importance of particular trajectories that come into grazing contact with the fluid layer or with the sample surface.

4. To precisely quantify the power dissipated due to interactions with the capillary fluid layer and to relate this to the cantilever dynamics.

5. To determine the global domains of attraction of steady oscillating solutions. This will yield general insights which could be useful in the development control strategies.

1.3 Outline

The following outlines a summary of the presented research:

Chapter 2: We discuss in detail the numerical approaches used to study the nonlinear dynamics of the AFM. Forward-time simulation and pseudo-arclength continuation methods are discussed for hybrid dynamical systems. A simple example oscillator is studied using both numerical approaches.

Chapter 3: We discuss a lumped mass spring model of the AFM that includes interaction forces consisting of attractive, adhesive, repulsive, and capillary contributions. Both forward-time simulation and pseudo-arclength continuation methods are employed to find steady state
attractors and bifurcation diagrams.

Chapter 4: The energy dissipation in tapping mode atomic force microscopy operating under ambient conditions is investigated. The changes in energy dissipation and the maximum of dissipated energy are related to the cantilever dynamics.

Chapter 5: Using forward-time simulation for a large set of initial conditions, we compute two-dimensional projections of the basins of steady oscillating periodic solutions. We calculate and explore these basins over a range of equilibrium cantilever separations.

Chapter 6: We present conclusions and suggestions for future research.
Chapter 2

Numerical Approaches for Hybrid Dynamical Systems

In this chapter\(^1\), we introduce the dynamical system theory, numerical approaches, and terminology used throughout this dissertation. In particular, we define precisely what is meant by a hybrid dynamical system and discuss how we have tailored forward-time simulation and numerical continuation for use with these systems. We do this by exploring a mass-spring-damper configuration as a simple example of a hybrid dynamical system [47]. This chapter is tutorial in nature and information has been collected from a variety of references.

---

2.1 Forward-Time Simulation

2.1.1 Vector Fields

Forward-time simulation (alternatively direct numerical simulation) of a dynamical system corresponds to the approximation of the solution to a system of first-order ordinary differential equations as a function of time. It is convenient to collect the unknown values of the components of the solution in a column array, denoted by \( \mathbf{x} \) and referred to as the system state vector. The state vector is an element of a state space, such that every set of numerical values of the components of the state vector corresponds to a unique point in the state space. The number of components of \( \mathbf{x} \) denoted here by \( n \) is the dimension of the system.

A solution to the system of differential equations is represented by a curve \( \mathbf{x}(t) \) of values of the state vector parametrized by the independent variable \( t \) representing time. Indeed, the system of differential equations can then be written in the form

\[
\frac{d}{dt} \mathbf{x}(t) = f(\mathbf{x}(t), t),
\]

where the function \( f \) (evaluated at a point in state space and a time \( t \)) is known as the vector field. If \( f \) does not depend explicitly on \( t \), the corresponding dynamical system is said to be autonomous. In the case of an autonomous vector field, the rate of change of the state vector depends only on the current value of the state vector and not on the current time. Indeed, if \( \mathbf{\tilde{x}}(t) \) is a solution to Eq. (2.1) in the case of an autonomous vector field then \( \mathbf{\tilde{x}}(t) = \mathbf{\tilde{x}}(t + \tau) \) is also a solution for any constant \( \tau \). Thus, the solution in the case of an autonomous vector field depends only on elapsed time and not on absolute time.
In the case of continuously differentiable autonomous vector fields, there is a unique solution curve through every point in state space, although solutions on each such curve may not be defined for all values of $t$. In this case, it is possible to define a function $\Phi(x_0, t)$ known as the **flow** that represents the value of the state vector along a solution curve based at a point $x_0$ after an elapsed time $t$. It follows that,

$$\frac{d}{dt} \Phi(x_0, t) = f(\Phi(x_0, t)), \quad (2.2)$$

and,

$$\Phi(x_0, 0) = x_0. \quad (2.3)$$

Keeping $x$ fixed and varying $t$ then traces the solution curve through the point $x$ (Figure 2.1). Similarly, keeping $t$ fixed and varying $x$ corresponds to traversing a family of solution curves. By the uniqueness of solution curves, $\Phi(x_1, t_1) = \Phi(x_2, t_2)$ only if $x_1$ and $x_2$ lie on the same solution curve, in which case,

$$x_2 = \Phi(x_1, t_1 - t_2) \quad (2.4)$$

and,

$$x_1 = \Phi(x_2, t_2 - t_1). \quad (2.5)$$

### 2.1.2 Hybrid Dynamical Systems

It is often the case that models of the time evolution of a physical system consist of a collection of distinct systems of differential equations with conditions for when to switch
between them. Such systems are examples of hybrid dynamical systems. For example, in an
electronic circuit, two distinct systems of differential equations govern the dynamics in the
case when a switch is in the on position and when the switch is in the off position. It is
typically the case that the switching between the two systems of equations is governed by
one or several scalar-valued discontinuity trigger functions $h$ (also called event functions)
which may be functions of the state vector as well as time. Hybrid dynamical systems for
which the vector fields and discontinuity trigger functions do not depend explicitly on time
are said to be autonomous.

In the forward simulation of an autonomous hybrid dynamical systems, a complete descrip-
tion of the dynamics includes not simply the state vector, but also a value of a discrete index
variable that indicates which vector field governs the system at the present time. Thus, for
example, given initial conditions $x_0$ and $i_0$ for the state vector and the index variable, a
numerical approximation is sought of the solution to the initial-value problem,

\[
\frac{d}{dt} x(t) = f_{i_0}(x(t)) \\
x(t_0) = x_0
\]  

(2.6) (2.7)
until such a time \( t = t_1 \) that the value of one of the discontinuity trigger functions \( h \) changes sign from positive to negative, i.e., such that,

\[
h(x_1) = 0, \tag{2.8}
\]

and,

\[
\left. \frac{d}{dt} h(x(t)) \right|_{t=t_1} = h_x(x_1) \cdot f_{u_i}(x_1) < 0, \tag{2.9}
\]

where \( x_1 = x(t_1) \), a subscripted comma is our notation to indicate differentiation, and the inequality in the last expression follows from the chain rule of differentiation. Here, \( h_x \) is the \( 1 \times n \) row matrix of partial derivatives of the function \( h \) with respect to the components of its argument and \( \cdot \) simply indicates matrix multiplication. At this time, the value of the index variable is changed to \( j_1 \) according to some rule associated with the discontinuity trigger function. The simulation is then begun again to seek a numerical approximation of the solution to the initial-value problem,

\[
\frac{d}{dt} x(t) = f_{u_i}(x(t)),
\]

\[
x(t_1) = x_1, \tag{2.10}
\]

while monitoring zero-crossings of any relevant discontinuity trigger functions. The case of a single vector field considered at the beginning of this section is clearly a special case of a hybrid dynamical system, for which the index variable has a single value.

From this discussion, it is clear that the forward simulation of a hybrid dynamical system depends uniquely on the initial state vector and index variable. Thus, rather than recording
the entire solution curve, it suffices to record the value of the state vector and index variable at discrete moments in time. For example, if the sampling of a solution curve over arbitrary lengths of time results in a discrete number of state-space points, then the system behavior is periodic in time.

A convenient way to accomplish the proposed sampling is to introduce a sampling trigger function $h$, such that sampling occurs at a moment when the trigger function changes sign from positive to negative. Assuming that no solution curve of interest reaches a point where $h$ has a local minimum value of zero, the sampling is known as Poincaré sampling. In this case, it is convenient to introduce the Poincaré map $P$ as a function that maps a sampled state vector and index variable to the subsequent sampled state vector and index variable. For a periodic orbit, some iterate of the Poincaré map then has a fixed point, i.e., a combination of a state vector and index variable for which some later sample of the solution curve yields the identical value of the state vector and index variable.
2.1.3 An Example Oscillator

An example of an autonomous hybrid dynamical system is the oscillator shown schematically in Figure 2.3. Here, the mass $m$ moves under the influence of a linear spring with stiffness $k$, a viscous damper with damping coefficient $c$, and an additional interaction force $F_{\text{int}}$ between the mass as its environment that depends on the excursion $q$ of the mass relative to the undeformed length of the spring. Excitation of the oscillator is achieved through the imposition of a sinusoidally varying force with amplitude $F_d$ and angular frequency $\omega_d$.

![Figure 2.3: A schematic of an example oscillator. Here, the interaction force $F_{\text{int}}$ captures the onset of the additional restoring force for $q \geq d_0$.](image)

As suggested by Figure 2.3, two distinct regimes of interactions between the mass and its environment will be considered corresponding to the absence or presence of an elastic force resulting from the deformations of an additional spring with stiffness $K$ and zero load when $q \leq d_0$ for some constant $d_0$. For excursions in excess of $d_0$ this description thus results in an overall hardening spring response.

The equation governing the motion of the oscillator is given by,

\[
\begin{align*}
  m\ddot{q} + c\dot{q} + kq &= F_d \cos \omega_d t, & q < d_0 \\
  m\ddot{q} + c\dot{q} + kq &= F_d \cos \omega_d t - K(q - d_0), & q \geq d_0,
\end{align*}
\]

(2.12)
with driving force $F_d \cos \omega_d t$, position $q$, velocity $\dot{q}$, distance from the left wall to the bumper $d_0$, and the instantaneous separation between the mass and bumper $d = d_0 - q$. It is often useful to talk about the quality factor $Q$ of the oscillator which is simply $Q = m \omega_0 / c$ where $\omega_0$ is the resonant frequency of the mass-spring-damper. Since the mass-spring-damper is driven at its resonant frequency $\omega_d = \omega_0$.

The dynamics of the oscillator may be described through the introduction of a state vector

$$
\mathbf{x} = \begin{pmatrix} x_1 \\ x_2 \\ x_3 \end{pmatrix} \overset{\text{def}}{=} \begin{pmatrix} q \\ \dot{q} \\ \omega_d t \mod 2\pi \end{pmatrix}
$$

and an index variable $i$, such that

$$
f_i (\mathbf{x}) = \begin{pmatrix} x_2 \\ \frac{1}{m} (F_d \cos x_3 - cx_2 - k x_1 + F_{\text{int}} (x_1, i)) \\ \omega_d \end{pmatrix},
$$

where

$$
F_{\text{int}} (q, i) = \begin{cases} 0 & i = 0 \\ -K (q - d_0) & i = 1 \end{cases}
$$

and $x_3$ corresponds to the instantaneous phase of the excitation.

Table 2.1 shows the discrete changes in the value of the index variable that occur as a result of zero-crossings of certain discontinuity trigger functions from positive to negative values. As $F_{\text{int}} = 0$ when $q = d_0$, the variations of the interaction force are continuous across transitions between $i_b = 0$ and $i_a = 1$ and vice versa (although the gradient of the interaction force is
discontinuous across these transitions).

\[
\begin{array}{ccc}
i_b & i_a & 0 \quad 1 \\
0 & d_0 - x_1 \\
1 & x_1 - d_0
\end{array}
\]

Table 2.1: Each entry in the table represents the discontinuity trigger function for which a zero-crossing from positive to negative results in a discrete change in the index variable from its value \(i_b\) before the triggered event and its value \(i_a\) after the triggered event.

Now consider the sampling trigger function \(h(x) = x_2\), such that a zero value of \(h\) corresponds to \(x_2 = \dot{q} = 0\), i.e., zero translational velocity of the oscillating mass. Denote by \(x^* = x(t^*)\) a state-space point on a solution curve, such that \(h(x^*) = 0\) and \(h(x(t)) > 0\) for \(t < t^*\). Then, sampling occurs provided that

\[
\frac{d}{dt} h(x(t)) \bigg|_{t=t^*} = h_x(x^*) \cdot f_i(x^*) = \dot{x}_2 = \ddot{q} < 0,
\]

i.e., provided that the acceleration is negative at the moment when the speed equals zero (after having been positive). Thus, a record of the sampled values of the state vector corresponds to values of \(q\) and \(\omega_dt \mod 2\pi\) at moments when the excursion of the mass achieves a local maximum. In the case of a periodic response of the oscillator with period equal to that of the excitation, the sampled value of \(\omega_dt \mod 2\pi\) equals the phase lag of the response relative to the excitation (which achieves its maximum value at \(\omega_dt \mod 2\pi = 0\)).

As an example of the results of forward-time simulation and Poincaré sampling, Figure 2.4 shows a finite solution segment of the example oscillator with parameter values given in Table 2.2. Here, forward-time simulation was implemented using Matlab’s ode integrators and the corresponding event-handling functionality.
Table 2.2: Values of the system parameters used in the numerical simulations of the example oscillator.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mass</td>
<td>$m$</td>
<td>1 kg</td>
</tr>
<tr>
<td>Stiffness</td>
<td>$k$</td>
<td>1 N/m</td>
</tr>
<tr>
<td>Damping coefficient</td>
<td>$c$</td>
<td>1 kg/s</td>
</tr>
<tr>
<td>Interaction stiffness</td>
<td>$K$</td>
<td>100 N/m</td>
</tr>
<tr>
<td>Excitation magnitude</td>
<td>$F_d$</td>
<td>6 N</td>
</tr>
<tr>
<td>Excitation frequency</td>
<td>$\omega_d$</td>
<td>1 rad/s</td>
</tr>
</tbody>
</table>

We would like to be clear by what is meant by the phase $\phi$ and how it is calculated using $x_3$. When $x_3$ is evaluated at the point where the mass has zero velocity it yields the phase of the oscillating mass relative to the phase of the driving force. More specifically, given a driving force $F_0 = F_d \cos(\omega_d t)$ that is oscillating at the resonant frequency of the mass-spring-damper, the dynamics of the mass will be at the same frequency but with a possible phase shift, $q = A \cos(\omega_d t - \phi)$, \hspace{1cm} (2.17)

where $A$ is some constant amplitude. When the velocity is evaluated at this position its value is $\dot{q} = A\omega_d \sin(\omega_d t - \phi) = 0$ which is only possible when $\omega_d t = \phi$ or equivalently $x_3 = \phi$.

This approach for determining the phase is very convenient because it is a direct result of our calculations and does not require any further steps involving curve fitting our, spectral analysis, etc.

Each new simulation is begun with initial conditions given by the steady oscillations of the previous simulation. Consider the last oscillation calculated from a simulation at some value of $d_0^n$. Let us denote the values of the state vector when the mass is at its furthest point from the surface during this final oscillation by $x^n$. The initial conditions for the $n+1$ simulation at the next separation $d_0^{n+1} = d_0^n + \Delta d_0$ where $\Delta d_0$ is the chosen step size for changes in
Figure 2.4: A sample periodic trajectory of the example oscillator with $d_0 = 4$ m. Here, the dots refer to zero-crossings of the event functions corresponding to the onset and termination of interactions with the additional linear spring and the Poincaré sampling trigger function. The dashed line refers to the instantaneous change in $x_3$ from $2\pi$ to 0 that results from its definition.

Separation is then $x^{n+1} = x^n$. This is a choice we made for the initial conditions and there are many other possibilities. When multiple solutions are near one another, just by using different initial conditions one can trace a different path through the solution space.
2.2 Linearized Dynamics

2.2.1 The Variational Equations

Given the interpretation of the flow function, it follows that $\Phi (x_0 + dx, t)$ describes the value of the state vector along a solution curve based at a point $x_0 + dx$ after an elapsed time $t$. If $\|dx\| \ll 1$, i.e., if the deviation from $x$ is sufficiently small, it is possible to approximate $\Phi (x_0 + dx, t)$ by its value on the solution curve based at $x_0$ plus a small correction of the same order of magnitude as $dx$. In particular, from Taylor’s theorem it follows that

$$\Phi (x_0 + dx, t) \approx \Phi (x_0, t) + \Phi_x (x_0, t) \cdot dx,$$  \hspace{1cm} (2.18)

where $\Phi_x (x_0, t)$ is an $n \times n$ matrix whose entries are the partial derivatives of the components of the flow function with respect to the components of the first argument for constant $t$ evaluated at the point $x_0$. Put differently, the entries of $\Phi_x (x_0, t)$ consist of the rate of change of the components of the state-space point $\Phi (x_0, t)$ with respect to changes in the components of the initial state vector $x_0$. The Jacobian matrix $\Phi_x (x_0, t)$ then describes the sensitivity of the final condition to changes in the initial condition.

Given a certain initial condition $x_0$, the Jacobian $\Phi_x (x_0, t)$ may be approximated as a function of time by direct numerical simulation of the initial-value problem

$$\frac{\partial}{\partial t} \Phi_x (x_0, t) = f_x (\Phi (x_0, t)) \cdot \Phi_x (x_0, t),$$

$$\Phi_x (x_0, 0) = I,$$  \hspace{1cm} (2.19)
obtained directly from differentiation with respect to the initial condition of Eqs. (2.2-2.3). Here, \( f_x(\Phi(x_0, t)) \) is a \( n \times n \) matrix whose entries are the partial derivatives of the components of the vector field with respect to the components of its argument evaluated at the point \( \Phi(x_0, t) \) along the trajectory through \( x_0 \) and \( I \) denotes the \( n \times n \) identity matrix. In practice, forward simulation of the differential equations given by Eqs. (2.19), known as the first variational equations, is performed at the same time as that of the original differential equations Eq. (2.2).

### 2.2.2 Hybrid Dynamical Systems

In a hybrid dynamical system, the value of the state vector along a solution curve based at a point \( x_0 + dx \) after an elapsed time \( t \) cannot directly be obtained from the solution to the variational equations. Suppose, for example, that the solution curve based at \( x_0 \) with initial value \( i_0 \) of the index variable reaches a zero-crossing of the discontinuity trigger function \( h \) at time \( t_1 \) at a point \( x_1 = \Phi_{i_0}(x_0, t_1) \), see Figure 2.2. Suppose, moreover, that subsequent to the zero-crossing the index variable changes to \( i_1 \) and the system evolves until time \( t_2 \) at a point \( x_2 = \Phi_{i_1}(x_1, t_2 - t_1) \) without any further zero crossings. The solution \( \Phi_{i_0,x}(x_0, t) \) to the variational equation corresponding to \( i = i_0 \) then describes the rate of change of the components of the state-space point \( \Phi_{i_0}(x_0, t) \) at time \( t \) with respect to changes in the components of \( x_0 \). This ignores, however, the possibility that nearby solution curves will have reached the zero-crossing of the discontinuity trigger function \( h \) prior to \( t \). Indeed, the time elapsed from a given initial condition to the subsequent zero crossing depends in a nonlinear way on the initial condition.

The \textit{implicit function theorem} affords a rigorous mathematical technique for determining
under what conditions nearby solution curves will reach the zero-crossing of the discontinuity trigger function $h$ at a unique time close to $t_1$. Provided that these conditions are satisfied, the theorem further provides an approximation for the time of zero-crossing as well as of the state-space point at the moment of the zero-crossing as a function of the initial condition. Specifically, consider the function,

$$F(x, t) \overset{\text{def}}{=} h(\Phi_{i_0}(x, t)).$$

(2.20)

If a time $\tau$ can be found, given $x$, such that

$$F(x, \tau) = 0$$

(2.21)

and

$$\frac{d}{dt} F(x, \tau) < 0,$$

(2.22)

then $\Phi_{i_0}(x, t)$ corresponds to the state-space point at the moment of a zero-crossing.

From the assumptions made above,

$$F(x_0, t_1) = 0$$

(2.23)

and

$$\frac{d}{dt} F(x_0, t_1) < 0.$$  

(2.24)

The implicit-function theorem now implies that for every $x \approx x_0$, there exists a unique time $\tau(x) \approx \tau(x_0) = t_1$ for which the above conditions hold, i.e., a unique time for the
corresponding zero-crossing. Implicit differentiation of the equality

\[ F(x, \tau(x)) = 0 \quad (2.25) \]

with respect to \( x \) and evaluated at \( x_0 \) then yields

\[ F_x(x_0, \tau(x_0)) + F_t(x_0, \tau(x_0)) \tau_x(x_0) = 0. \quad (2.26) \]

Solving this for \( \tau_x(x_0) \) and substitution from the definition of \( F \), it follows that

\[
\tau_x(x_0) = - \frac{F_x(x_0, \tau(x_0))}{F_t(x_0, \tau(x_0))} = - \frac{h_x(\Phi_i(x_0, \tau(x_0))) \cdot \Phi_i x(x_0, \tau(x_0))}{h_x(\Phi_i(x_0, \tau(x_0))) \cdot f_i(x_0, \tau(x_0))} = - \frac{h_x(x_1) \cdot \Phi_i x(x_0, t_1)}{h_x(x_1) \cdot f_i(x_1)}. \quad (2.27)
\]

From Taylor’s theorem it follows that, for \( \|dx\| \ll 1 \),

\[
\Phi_i(x_0 + dx, \tau(x_0 + dx)) \approx x_1 + [\Phi_i x(x_0, t_1) + f_i(x_1, \tau_x(x_0))] \cdot dx
\]

\[
= x_1 + \left[ I - \frac{f_i(x_1) \cdot h_x(x_1)}{h_x(x_1) \cdot f_i(x_1)} \right] \cdot \Phi_i x(x_0, t_1) \cdot dx \quad (2.28)
\]

for the approximate state-space point corresponding to the zero-crossing along the solution curve based at \( x_0 + dx \).

The state along the solution curve based at \( x_0 + dx \) at time \( t_2 \) is now obtained from the expression,
\[ \Phi_{i_1} (\Phi_{i_0} (x_0 + d\mathbf{x}, \tau (x_0 + d\mathbf{x})), t_2 - \tau (x_0 + d\mathbf{x})) \approx \]

\[ \approx x_2 + \left[ \Phi_{i_1,x} (x_1, t_2 - t_1) \cdot \left[ \Phi_{i_0,x} (x_0, t_1) + f_{i_0} (x_1) \cdot \tau_x (x_0) \right] \right] \cdot d\mathbf{x} \]

\[ = x_2 + \Phi_{i_1,x} (x_1, t_2 - t_1) \cdot \left[ I + \left( \frac{f_{i_1} (x_1) - f_{i_0} (x_1)}{h_x (x_1) \cdot f_{i_0} (x_1)} \right) \cdot \Phi_{i_0,x} (x_0, t_1) \right] \cdot d\mathbf{x}, \]  

(2.29)

where we used the identity

\[ f_{i_1} (x_2) = \Phi_{i_1,x} (x_1, t_2 - t_1) \cdot f_{i_1} (x_1) \]  

(2.30)

that follows from the definition of the flow. In conclusion, the rate of change of the final condition at time \( t_2 \) with respect to changes in the initial condition \( x_0 \) is given by the matrix,

\[ \Phi_{i_1,x} (x_1, t_2 - t_1) \cdot \left[ I + \left( \frac{f_{i_1} (x_1) - f_{i_0} (x_1)}{h_x (x_1) \cdot f_{i_0} (x_1)} \right) \right] \cdot \Phi_{i_0,x} (x_0, t_1) . \]  

(2.31)

The present discussion carries over directly to the case of a zero-crossing for a sampling trigger function \( h_P \). Suppose, for example, that \( x_0 \) and \( x_2 \) on the solution curve described above are two consecutive zero-crossings for the sampling trigger function \( h_P \), i.e., such that
\( \mathbf{P} (x_0) = x_2 \) for the corresponding Poincaré map. Then

\[
\mathbf{P}_x (x_0) = \left[ I - \frac{f_{i_1} (x_2)}{h_{P,x} (x_2)} \right] \cdot \Phi_{i_1,x} (x_1, t_2 - t_1) \cdot \left[ I + \frac{(f_{i_1} (x_1) - f_{i_0} (x_1))}{h_{i,x} (x_1)} \right] \cdot \Phi_{i_0,x} (x_0, t_1) \tag{2.32}
\]

as follows by composition of the expressions derived above. In the special case that \( x_2 = x_0 \), i.e., when sampling a periodic function, the sensitivity of the sampled data to perturbations in the initial state vector follows directly from \( \mathbf{P}_x (x_0) \). Indeed, as long as all the eigenvalues of \( \mathbf{P}_x (x_0) \) lie within the unit circle, then all solutions based at nearby initial conditions decay exponentially fast toward the periodic solution to \( x_0 \) which is then said to be \textit{asymptotically stable}. If, instead, at least one eigenvalue lies outside the unit circle, then the periodic solution is \textit{unstable} and it is possible to find solutions based at arbitrarily close initial conditions that deviate from the periodic solution in forward time. These conclusions may be directly extended to arbitrary iterates of \( \mathbf{P}_x (x_0) \) and more complicated sequences of switchings between different vector fields.

### 2.2.3 The Example Oscillator

In the case of the example oscillator described above with \( i = 0 \),

\[
f_{0,x} (x) = \begin{pmatrix} 0 & 1 & 0 \\ -k/m & -c/m & -F_d \sin x_3/m \\ 0 & 0 & 0 \end{pmatrix} \tag{2.33}
\]
Let
\[
\Phi_{0,x}(x_0, t) \stackrel{\text{def}}{=} \begin{pmatrix}
    x_4(t) & x_5(t) & x_6(t) \\
    x_7(t) & x_8(t) & x_9(t) \\
    x_{10}(t) & x_{11}(t) & x_{12}(t)
\end{pmatrix}.
\] (2.34)

Then, the variational equations correspond to the system of differential equations
\[
\frac{d}{dt} \begin{pmatrix}
    x_4(t) & x_5(t) & x_6(t) \\
    x_7(t) & x_8(t) & x_9(t) \\
    x_{10}(t) & x_{11}(t) & x_{12}(t)
\end{pmatrix} = \begin{pmatrix}
    0 & 1 & 0 \\
    -k/m & -c/m & -F_d \sin (x_3/m) \\
    0 & 0 & 0
\end{pmatrix} \cdot \begin{pmatrix}
    x_4(t) & x_5(t) & x_6(t) \\
    x_7(t) & x_8(t) & x_9(t) \\
    x_{10}(t) & x_{11}(t) & x_{12}(t)
\end{pmatrix}.
\] (2.35)
With \( h_0(x) = d_0 - x_1 \) it follows, for example, that at a point \( x_1 \), at which \( h_0(x_1) = 0 \),

\[
I - \frac{f_0(x_1) \cdot h_{0,x}(x_2)}{h_{0,x}(x_1) \cdot f_0(x_1)} = \begin{pmatrix}
1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 1 \\
\end{pmatrix} - \begin{pmatrix}
x_2 \\
\frac{1}{m}(F_d \cos x_3 - cx_2 - kx_1) \\
\omega_d \\
\end{pmatrix} \cdot \begin{pmatrix}
-1 & 0 & 0 \\
\end{pmatrix}
\]

\[
= \begin{pmatrix}
0 & 0 & 0 \\
\frac{1}{m x_2} (F_d \cos x_3 - cx_2 - kx_1) & 1 & 0 \\
\omega_d / x_2 & 0 & 1 \\
\end{pmatrix}.
\]

### 2.3 The Numerical Continuation of Periodic Solutions

Forward simulation is an important tool in the analysis of the dynamics of the AFM. Using a lumped model for the AFM results in a second order nonlinear ordinary differential equation governing the dynamics of the cantilever. The study of nonlinear ordinary equations is an actively growing area of research with many powerful and insightful results and techniques. In studying the AFM we will apply numerical continuation to follow periodic solutions and in the following we briefly discuss the basic and important ideas that are required. Here is a brief outline of the approach:

1. Formulate the governing equation as a collection of autonomous systems of first order ordinary differential equations. Each system of equations describes the dynamics of
the cantilever during a distinct phase of its motion.

2. Formulate a collection of *discontinuity trigger functions* whose zero-crossings trigger the termination of one phase of motion and the onset of another.

3. Define an interesting Poincaré section (event surface) in state space in which to study the dynamics. For illustration consider the surface in state space for which the cantilever is at a position furthest from the surface during a period.

4. Since the dynamics are periodic the solution will be a fixed point on the Poincaré section. Furthermore, one can define a Poincaré map that maps the dynamics from one intersection with the Poincaré section to the next.

5. Define a function that vanishes when evaluated at the intersection with the Poincaré section for the current parameter value. Using this function and when certain conditions are met (discussed further below) the implicit function theorem guarantees the existence and uniqueness of a solution for a small change in the parameter [49].

6. Use a root-finding scheme (such as Newton-Raphson) to find a solution to the dynamics at the new value of the parameter. Therefore, the solution can be followed through parameter space as the parameter is varied. For a standard continuation approach one could use a parameter from the system such as the equilibrium separation between the cantilever and surface. However, we will find it useful to implement pseudo-arclength continuation.

In what follows we provide the details for these steps. Consider a continuous-time system of ordinary differential equations that has been replaced by a system of $n$ first order ordinary
differential equations where the time $t$ has also been made a state variable. This autonomous system is then,

$$\dot{x} = f(x; \mu),$$

(2.37)

where $x$ is a $n$-dimensional state vector, $f$ is the $n$-dimensional vector field, $\mu$ is a $m$-dimensional vector of system control parameters, and a dot indicates time differentiation.

Using numerical continuation we will determine the periodic solution to this system of equations as a particular parameter of the system $p$ is changed. In proceeding it will be useful to think of the vector field as a function of the state vector and the parameter $p$ that is to be varied.

We are interested in the sensitivity of the periodic dynamics to small changes of $x$ away from the point $x_0$ (a fixed point on the Poincaré section). This is given by the Jacobian of the Poincaré map evaluated at the point $x_0$ which is,

$$P_{x}(x_0) = \Phi_{,t}(x, \tau(x)) \cdot \tau_x(x_0) + \Phi_{,x}(x_0, \tau(x)).$$

(2.38)

Substituting in for $\tau_x(x_0)$ yields,

$$P_{x}(x_0) = [I - \frac{f(x_0) \cdot h_x(x_0)}{h_x(x_0) \cdot f(x_0)}] \cdot \Phi_{,x}(x_0, \tau(x_0))$$

(2.39)

where $\Phi_{,x}(x_0, \tau(x_0))$ is the solution of the first variational equations.
2.3.1 Pseudo-arclength Continuation of Periodic Solutions

In the case of periodic solutions, it is possible to implement a continuation analysis method that avoids the need to simulate past the initial transients and is independent of the existence of a basin of attraction. Consider, for example, a dynamical system with a single vector field and suppose that a Poincaré sampling trigger function $h_P$ has been found, such that $x_0$ is a fixed point of the corresponding Poincaré map for some choice of values $\mu_0$ of the vector of system parameters. Moreover, suppose that the nullspace of the matrix

$$
\begin{pmatrix}
P_{x,x}(x_0, \mu_0) - I & P_{\mu}(x_0, \mu_0)
\end{pmatrix}
$$

is one-dimensional and spanned by the vector

$$
\tau = \begin{pmatrix} t_x \\ t_\mu \end{pmatrix}, \quad (2.40)
$$

i.e., that

$$
\begin{pmatrix}
P_{x,x}(x_0, \mu_0) - I & P_{\mu}(x_0, \mu_0)
\end{pmatrix} \cdot \begin{pmatrix} \alpha \\ \beta \end{pmatrix} = 0 \quad (2.41)
$$

if and only if

$$
\begin{pmatrix} \alpha \\ \beta \end{pmatrix} = k\tau. \quad (2.42)
$$

It follows that $x = x_0$, $\mu = \mu_0$, and $\lambda = 0$ is a solution to the equation

$$
F(x, \mu, \lambda) \overset{def}{=} \begin{pmatrix}
P(x, \mu) - x \\ (x - x_0) \cdot \tau - \lambda
\end{pmatrix} = \begin{pmatrix} 0 \\ 0 \end{pmatrix} \quad (2.43)
$$
and that
\[
\mathbf{F}(\mathbf{x}, \mu) (\mathbf{x}_0, \mu_0, 0) = \begin{pmatrix}
P_{\mathbf{x}} (\mathbf{x}_0, \mu_0) - \mathbf{I} & P_{\mu} (\mathbf{x}_0, \mu_0) \\
\mathbf{t}_{\mathbf{x}} & \mathbf{t}_{\mu}
\end{pmatrix}
\]
(2.44)
is invertible. By the implicit function theorem, it follows that for every \( \lambda \approx 0 \), there exists a unique fixed point \( \mathbf{x}(\lambda) \approx \mathbf{x}(0) = \mathbf{x}_0 \) corresponding to the value \( \mu(\lambda) \approx \mu(0) = \mu_0 \) of the vector of system parameters.

Having established the persistence of a fixed point (and corresponding periodic solution) under variations in the pseudo-arclength parameter \( \lambda \), continuation is now achieved by implementing a Newton-Raphson algorithm to trace the branch of periodic solutions parametrized by \( \lambda \) [49, 50].

### 2.3.2 Stability of Solutions

According to the Hartman-Grobman theorem [49], the fixed point \( \mathbf{x}_{\text{ref}} \) is stable if all of the eigenvalues of \( P_{\mathbf{x}} (\mathbf{x}_{\text{ref}}) \) lie within the unit circle in the complex plane (i.e. have magnitude less than unity). If at least one eigenvalue lies outside the unit circle, the fixed point is unstable. The crossing of an eigenvalue through the unit circle is typically associated with a bifurcation in system characteristics, for example one such possibility is the appearance of periodic trajectories with half the frequency (period doubling).
2.3.3 The Example Oscillator

For the example oscillator, a periodic steady-state attractor for which \( i \) is identically equal to 0 can be found for sufficiently large \( d_0 \) given by

\[
q(t) = A_0 \cos(\omega_d t - \phi),
\]

where

\[
A_0 = \frac{F_d}{m \sqrt{(k - m \omega_d^2)^2 + (c \omega_d)^2}}
\]

is the amplitude of oscillation and the phase lag \( \phi \) satisfies the equation

\[
tan \phi = \frac{c \omega_d}{k - m \omega_d^2}.
\]

In particular, it follows that this steady-state attractor persists with a finite basin of attraction as long as \( d_0 > A_0 \). On the other hand, for \( d_0 < A_0 \) no steady-state attractor will exist for which \( i \) remains equal to 0.

Both forward-time simulation and pseudo-arclength continuation may be employed to trace this steady-state attractor as \( d_0 \) is decreased below the critical value \( A_0 \). Figures 2.5 and 2.6 show the sampled values of \( q \) and \( \omega_d t \mod 2\pi \), respectively, as obtained by forward-time simulation (circles) and pseudo-arclength continuation (solid line) as \( d_0 \) is decreased past \( A_0 \) for the parameter values shown in Table 2.2.

In particular, in the case of forward-time simulation, continuation is initiated at \( d_0 = A_0 \) with initial conditions obtained from the exact solution above. After each change \( \Delta d_0 = -0.2 \) m, the simulation is performed for 50 cycles of the excitation and the last five sampled values
Figure 2.5: (color) Sampled values of $q$ at points of maximum excursion obtained during continuation of the periodic steady-state attractor for the example oscillator using both forward-time simulation (red, circles) and pseudo-arclength continuation (blue, line) under variations in $d_0$. Periodic trajectories represent two distinct regimes of interactions between the mass and its environment. $i = 0$ shows the portion of the trajectory that does not interact with the additional spring with stiffness $K$ and $i = 1$ shows the excursions in excess of $d_0$ which results in an overall hardening spring response.

of the state vector are included in the figure. In contrast, the pseudo-arclength continuation is implemented using an adaptive step algorithm that seeks to maximize $\lambda$ (with an upper bound of 0.2) while achieving convergence of the Newton scheme within five iterations.

It is clear from the figures that the periodic steady-state attractor persists past the critical value $A_0$ but that its oscillation amplitude and phase lag decrease relative to those found using the above formulae. Of particular interest are parameter values for which the periodic
Figure 2.6: (color) Sampled values of $\omega d t \mod 2\pi$ at points of maximum excursion obtained during continuation of the periodic steady-state attractor for the example oscillator using both forward-time simulation (red, circles) and pseudo-arclength continuation (blue, line) under variations in $d_0$.

trajectory satisfies the conditions

\begin{align}
    x_1(t^*) &= d_0, \\
    x_2(t^*) &= 0
\end{align}

for some $t = t^*$ corresponding to so-called grazing contact of the trajectory with the surface $x_1 = d_0$ plane in state space. In the mechanical model this corresponds to a zero-relative-velocity contact between the oscillating mass and the additional spring. As seen in the inserts in the figure, each value of $d_0$ corresponding to grazing contact separates a family of periodic trajectory with a distinct sequence of values for the index variable.
Chapter 3

The Nonlinear Dynamics of Tapping Mode Atomic Force Microscopy with Capillary Force Interactions

In this chapter\footnote{Significant portions of this chapter have been published in N. Hashemi, H. Dankowicz, and M.R. Paul, The nonlinear dynamics of tapping mode Atomic Force Microscopy with capillary force interactions, \textit{J. Appl. Phys.}, 103, 093512 (2008).}, the nonlinear dynamics of a tapping mode atomic force microscope has been studied using forward-time simulation and numerical pseudo-arclength continuation [19]. Atomic force microscopy (AFM) has been used extensively to study the surface topography of a wide variety of materials and has also proven capable of measuring intramolecular forces [2, 4, 5]. When operating in tapping mode, the cantilever probe makes intermittent contact with the sample and, as a result, can be expected to reduce sample destruction during measurement as compared to contact modes. Consequently, the tapping mode has been widely employed to study compliant materials such as polymers, biomaterials, and semicon-
ductors [4,20]. The interaction between the AFM probe tip and the sample is discontinuous or exhibits discontinuous changes in its rate of change with tip-sample separation, is hysteretic as in the presence of capillary interactions, and highly nonlinear. A substantial and growing literature is dedicated to low-complexity modeling of these interactions and a study of their implications to the probe dynamics [16–18,20,22–25].

Previous work has shown that during tapping mode operation, the AFM probe dynamics explore two distinct branches of stable oscillations as one varies the equilibrium separation between the cantilever and the sample surface [16, 20]. In the idealized absence of noise, transitions between these branches is found to be associated with the onset of repulsive tip-sample interactions when the cantilever came into contact with the sample surface [16], whereas premature transitions would be observed away from these conditions in the presence of noise. Zitzler et al. [17] explored the influence on these transitions of capillary forces, resulting from a thin layer of water on the sample and probe tip due to humidity in the surrounding air.

In this chapter we study in detail the AFM dynamics using the model of Zitzler et al. [17] by developing and using specialized numerical algorithms that carefully treat the discontinuous and hysteretic spatial dependence of the force interactions between the cantilever tip and sample surface. While the theoretical tools particular to such hybrid systems are not detailed here (but can be found in the references), the resultant formalism avoids the introduction of additional model assumptions at the stage of numerical implementation of a forward-simulation model. Moreover, the hybrid formulation is associated with a well-defined algorithm for evaluating the dynamic stability of periodic system responses, as well as with techniques for tracing such responses under parameter variations regardless of their
stability. In contrast to Zitzler et al. [17], the chapter therefore embraces the piecewise nature of the system definition and exploits this so as to plausibly explain complexities in the system response.

Hybrid dynamical systems form a natural backbone for the analysis of mechanical systems with impacts or dry friction (including conditions of stick or slip), electrical circuitry with nonlinear circuit elements, such as diodes and transistors, and biomolecular models with chemical switches, such as the mitotic halving of the cell mass (cf. [51]). In these cases, as with the application considered here, and in contrast to smooth models, the hybrid formalism affords a means to accurately resolve changes in the system response that are directly associated with rapid changes in the system state or the state of system interactions [52].

3.1 The Physical Model of the Tip Surface Interactions

The atomic force microscope is composed of an elastic cantilever whose dynamics varies due to complex force interactions $F_{ts}$ between the cantilever tip and the sample surface. Following [17], $F_{ts}$ is assumed to include attractive long-range van der Waals forces $F_v$, capillary contributions $F_c$ due to the presence of a liquid layer, and repulsive forces $F_r$ representing the onset of contact. The objective of this section is to briefly outline the important model assumptions and quantitative expressions for each of these interactions. The reader is referred to Ref. [17] for further details.

Let $d$ denote the instantaneous nominal tip-sample separation, i.e., the distance between the cantilever tip and a nominal reference plane associated with the sample surface. Assuming the interaction can be modelled as a sphere in close proximity to an infinite surface the van
der Waals forces are given by \( F_v = HR/6d^2 \) when \( d > a_0 \), where \( H \) is the Hamaker constant, \( R \) is the radius of curvature of the cantilever tip, and \( a_0 \) is the intermolecular constant. For separations \( d \leq a_0 \), the van der Waals force is assumed to be constant and equal to its value when \( d = a_0 \). The Derjaguin-Muller-Toporov (DMT) contact model [53] yields \( a_0 = (H/24\pi\gamma_{sv})^{1/2} \) where \( \gamma_{sv} \) is the surface energy of the tip and sample.

Contact between the cantilever tip and the sample surface, corresponding to \( d < a_0 \), is associated with a strong repulsive force. Again using DMT theory this repulsive interaction is given by \( F_r = -(4/3)E^sR^{1/2}(a_0 - d)^{3/2} \), where \( E^{s-1} = (1 - \nu_t^2)/E_t + (1 - \nu_s^2)/E_s \) and \( E_t, E_s, \nu_t, \nu_s \) are the Young’s modulus and Poisson ratio for the tip (\( t \)) and sample (\( s \)).

The capillary force arises from the interactions between thin films of water of depth \( h \) that cover the sample and cantilever tip due to ambient humidity [17,42,54–59]. As the separation \( d \) falls below a critical distance \( d_{on} = 2h \), a connective column of liquid is established. Upon retracting away from the surface the liquid column forms a meniscus and neck until eventually breaking as the separation increases beyond a critical distance \( d_{off} = V^{1/3} - V^{2/3}/5R \) where \( V \) is the meniscus volume. In the presence of the liquid column, the force interaction caused by the water layers can be modelled as \( F_c = 4\pi\gamma_wR/(1 + d/h) \) when \( d > a_0 \), where \( \gamma_w \) is the surface energy of water. For separations \( d \leq a_0 \), the capillary force is assumed to be constant and equal to its value when \( d = a_0 \).

During tapping-mode operation, a low-dimensional model reduction that provides a reasonably accurate description of the cantilever dynamics represents the cantilever by a lumped mass \( m \) that moves under the influence of a linear spring with stiffness \( k \), a viscous damper with damping coefficient \( c \), a harmonic driving with amplitude \( F_d \) and angular frequency \( \omega_d \) (here assumed to equal the natural frequency \( \omega_0 = \sqrt{k/m} \)), and the tip-sample interaction.
force $F_{ts}$ discussed above. A schematic of such a model is shown in Figure 3.1.

Figure 3.1: A schematic of the lumped-mass model used to represent an AFM cantilever oscillating near a sample surface. The cantilever has mass $m$, spring constant $k$, and damping coefficient $c$. The cantilever is driven with a force of magnitude $F_d$ and frequency $\omega_d$. The interaction force $F_{ts}$ captures the attractive, capillary, and repulsive interactions between the cantilever tip and the sample. The cantilever is shown at the equilibrium rest position which is at a distance $d_0$ from the sample surface. Also shown is the distance from the equilibrium rest position of the cantilever to the onset of the capillary force $d_0 - d_{on}$, contact with the sample surface $d_0 - a_0$, and the release of the capillary force $d_0 - d_{off}$. The specific parameter values used in our numerical investigation are listed in Table 3.2.

The corresponding equation of motion is now given by

$$m\ddot{q} + c\dot{q} + kq = F_d \cos \omega_d t + F_{ts},$$  \hspace{1cm} (3.1)

where $q = d_0 - d$ is the instantaneous displacement of the cantilever tip measured from the equilibrium tip position in the absence of external forces (i.e. for an unstretched spring) with positive values towards the sample surface, and $\dot{q}$ and $\ddot{q}$ are the instantaneous velocity and acceleration, respectively, of the cantilever tip.
3.2 Treating the AFM as a Hybrid Dynamical System

The objective of this section is to recast the physical model discussed above as a hybrid dynamical system supporting a rigorous simulation formalism, within which the bifurcation characteristics of the atomic-force microscope during tapping-mode operation may be carefully explored. The details of this can be found in Section 2.2 where we present the full analysis for a simple example oscillator.

Consider the three-dimensional continuous state vector $\mathbf{x}$ given by

\[
\mathbf{x} = \begin{pmatrix}
x_1 \\
x_2 \\
x_3
\end{pmatrix} = \begin{pmatrix}
q \\
\dot{q} \\
\omega d \mod 2\pi
\end{pmatrix}.
\] (3.2)

Here, $x_1$ and $x_2$ capture the mechanical state of the cantilever while $x_3$ describes the instantaneous phase of the periodic driving. In addition to the continuous state vector $\mathbf{x}$, a complete description of the instantaneous state of the tip dynamics requires knowledge of the state of the tip-sample interactions, particularly the specific combination of tip-sample interaction forces that are active at a given moment. For this purpose, let $i$ denote a discrete state variable, such that

\[
F_{ts}(q, i) = \begin{cases}
\frac{HR}{6(d_0-q)^2} & i = 0 \\
\frac{HR}{6(d_0-q)^2} + \frac{4\pi\gamma w R}{1+(d_0-q)/h} & i = 1 \\
\frac{HR}{6a_0^2} + \frac{4\pi\gamma w R}{1+a_0/h} - \frac{4}{3}E^* \sqrt{R}(a_0 - d_0 + q)^3/2 & i = 2
\end{cases}.
\] (3.3)

In particular, $i = 0$ corresponds to the state in which the tip-sample interaction force is...
entirely attractive and given by the van der Waals force. Similarly, $i = 1$ corresponds to
the state in which the tip-sample interaction force is a combination of attractive van der
Waals and capillary force contributions. Finally, $i = 2$ corresponds to the state in which the
tip-sample interaction force is a combination of two attractive constant contributions (that
equal the van der Waals and capillary force terms, respectively, when $d = a_0$) and a repulsive
force contribution.

The dynamics of the AFM cantilever during tapping-mode operation is captured through
a combination of continuous-in-time evolution of the continuous state vector $x$ as per the
autonomous system of first-order differential equations

$$
\dot{x} = f(x, i) = \begin{pmatrix} x_2 \\ m^{-1} (F_d \cos x_3 - cx_2 - kx_1 + F_{ts}(x_1, i)) \\ \omega_d \end{pmatrix}
$$

(3.4)

and discrete-in-time changes of the discrete state variable $i$ as per table 3.1. Specifically,

<table>
<thead>
<tr>
<th>$i_b$</th>
<th>$i_a$</th>
<th>$h$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>1</td>
<td>$d_0 - q - d_{on}$</td>
</tr>
<tr>
<td>1</td>
<td>0</td>
<td>$d_{off} - d_0 + q$</td>
</tr>
<tr>
<td>1</td>
<td>2</td>
<td>$d_0 - q - a_0$</td>
</tr>
<tr>
<td>2</td>
<td>1</td>
<td>$a_0 - d_0 + q$</td>
</tr>
</tbody>
</table>

Table 3.1: An exhaustive list of all possible non-degenerate transitions between tip-sample
interaction states as described by the index $i$ where $i_b$ is the value of the index before
the triggered event and $i_a$ is the value of the index after the triggered event. Also shown are
the event functions, $h$, for which a transversal zero-crossing from positive to negative values
triggers the indicated discrete change in the index variable.

suppose that $i = 0$ whenever $d_0 - q > d_{off}$ and $i = 2$ whenever $d_0 - q < a_0$. Each row of
table 3.1 then corresponds to a transition between two distinct combinations of tip-sample
interactions triggered by a transversal (i.e., non-tangential) zero-crossing of a characteristic event function from positive to negative values. In particular, let \( h(\mathbf{x}) \) be a function of the continuous state vector. Then, for a given value of the discrete state variable \( i \), such a transversal zero-crossing of the value of \( h \) along a system trajectory occurs at a time \( t = t^* \) provided that

\[
h(\mathbf{x}(t))|_{t=t^*} = h(\mathbf{x}(t^*)) = 0 \tag{3.5}
\]

and

\[
\frac{d}{dt} h(\mathbf{x}(t)) \bigg|_{t=t^*} = \partial_x h(\mathbf{x}(t^*)) \cdot \mathbf{f}(\mathbf{x}(t^*), i) < 0. \tag{3.6}
\]

Here, \( \partial_x h \) denotes the gradient of the function \( h \) with respect to the components of the state vector \( \mathbf{x} \) and the \( \cdot \) denotes matrix multiplication.

Table 3.1 gives an exhaustive list of all possible non-degenerate transitions between distinct combinations of tip-sample interactions and the associated event functions. In particular, the change between \( i_b = 0 \) and \( i_a = 1 \) is associated with a discontinuous jump of \( 4\pi \gamma w R/(1 + d_{on}/h) \) in the tip-sample interaction force corresponding to the onset of the capillary interactions. A similar discontinuous jump of \( 4\pi \gamma w R/(1 + d_{off}/h) \) in the tip-sample interaction force is associated with the transition between \( i_b = 1 \) and \( i_a = 0 \) and corresponds to the delayed rupture of the meniscus neck. In contrast, the transitions between \( i_b = 1 \) and \( i_a = 2 \) and vice versa are not associated with discontinuities in the tip-sample interaction force, although the gradient of the tip-sample interaction force is discontinuous across these transitions.

In order to accurately account for these transitions during forward-time numerical simulation, one proceeds in the following manner. Given consistent and non-degenerate initial conditions
\( x_0 \) and \( i_0 \), seek a numerical approximation for the solution to the initial-value problem

\[
\dot{x}(t) = f(x(t), i_0), \quad x(t_0) = x_0
\]  

(3.7)

until a time \( t = t_1 \) corresponding to a transversal zero-crossing of the value of one of the event functions \( h \) associated with \( i_b = i_0 \) from positive to negative. To continue beyond the corresponding transition, repeat the above construction after making the substitutions \( t_0 = t_1, x_0 = x(t_1), \) and \( i_0 = i_a \).

### 3.3 Numerical Results for a Tapping Mode AFM with Capillary Layer Interactions

The objective of this section is to numerically investigate the oscillatory dynamics of an atomic-force microscope cantilever driven by a single-frequency excitation of its clamped end for the parameter values listed in Table 3.2. To enable some reasonable comparison, we have chosen to use the values listed in [17] corresponding to a commercially available AFM that has a silicon tip tapping a silicon surface.

A typical experimental amplitude-separation curve would be obtained by graphing a representative measurement of the extent of the cantilever oscillations as the equilibrium position of the cantilever is brought toward the sample surface and then retracted, i.e., as \( d_0 \) is decreased and subsequently increased. Assuming that the resting time subsequent to each change in the equilibrium position is sufficiently long, the post-transient cantilever response can be identified with a steady-state attractor of the corresponding dynamical system. The
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spring constant</td>
<td>$k$</td>
<td>27.5 N/m</td>
</tr>
<tr>
<td>Quality factor</td>
<td>$Q$</td>
<td>400</td>
</tr>
<tr>
<td>Resonant frequency</td>
<td>$f_0$</td>
<td>280 KHz</td>
</tr>
<tr>
<td>Tip radius</td>
<td>$R$</td>
<td>20 nm</td>
</tr>
<tr>
<td>Intermolecular distance</td>
<td>$a_0$</td>
<td>0.103 nm</td>
</tr>
<tr>
<td>Water film thickness</td>
<td>$h$</td>
<td>0.2 nm</td>
</tr>
<tr>
<td>Capillary force turns on</td>
<td>$d_{on}$</td>
<td>0.4 nm</td>
</tr>
<tr>
<td>Capillary force turns off</td>
<td>$d_{off}$</td>
<td>2.32 nm</td>
</tr>
</tbody>
</table>

Table 3.2: Values of the parameters used in the numerical analysis of the AFM cantilever dynamics. The cantilever is driven at the resonant frequency $\omega_d = \omega_0 = 2\pi f_0$, the equivalent mass of the cantilever is $m = k/\omega_0^2$, and the damping is given by $c = m\omega_0/Q$. The Hamaker constant $H = 6.0 \times 10^{-20}$ J, the elastic modulus of the tip $E_t = 120$ GPa, the Poisson coefficient of the tip $\nu_t = 0.5$, the elastic modulus of the sample $E_s = 120$ GPa, the Poisson coefficient of the sample $\nu_s = 0.5$, the surface energy $\gamma_{sv} = 75$ mJ/m$^2$, and the surface energy of water $\gamma_w = 72$ mJ/m$^2$. For all of the results presented here we have chosen $F_d$ that yields a free amplitude of oscillation of the cantilever to be $A_0 = 30$ nm.

steady-state attractor observed subsequent to each change in equilibrium position is then a function of the basin of attraction within which lie the initial conditions obtained from the previously found attractor and, naturally, the influence of noise and uncertainty.

Both forward-time simulation and numerical continuation [60] may be employed to trace a periodic steady-state attractor as $d_0$ is varied. Numerical continuation is preferred due to its rapid rate of convergence, improved accuracy, and ability to continue the periodic trajectory even beyond bifurcation points characterized by a change in the linear stability. On the other hand, forward-time simulation becomes a necessary tool in the event that a branch of periodic trajectories terminates for some parameter value as we show below.

Figure 3.2 shows four branches of periodic steady-state solutions obtained using numerical continuation. The ordinate axis illustrates the minimum tip-sample distance during an oscillation (cf. Figure 3.1).
As expected, there exists a branch of periodic solutions (labeled 1 in Figure 3.2) that are independent of the sample position for sufficiently large equilibrium separations. This branch terminates at an equilibrium separation of $d_0 \approx 30.36$ nm, at which the corresponding periodic trajectory achieves \textit{simple grazing contact} with the water layer represented by the $x_1 = d_0 - d_{on}$ plane in state space. A further decrease in $d_0$ results in the onset of capillary interactions. Since the capillary force interaction persists until the trajectory crosses the $x_1 = d_0 - d_{off}$ plane in state space, the corresponding perturbation to the periodic trajectory is not small. As a result, no periodic trajectory with period equal to the drive period that includes capillary interactions is expected to emanate from this point.

![Figure 3.2: Branches of periodic solutions obtained using numerical pseudo-arclength continuation. The points labeled A-G correspond to periodic solutions that achieve grazing contact with a surface defined by an event function. Specifically, points A, B, C, and E represent grazing contact with the onset of capillary interactions given by the $x_1 = d_0 - d_{on}$ plane in state space (a dotted horizontal line is included to guide the eye). Points D, F, and G represent grazing contact with termination of the capillary force given by the $x_1 = d_0 - d_{off}$ plane in state space.](image-url)
To proceed beyond the onset of capillary force interactions, forward-time simulation is performed following the numerical approach discussed previously. The upper panel of Figure 3.3 shows the family of steady-state attractors obtained by initializing the forward time simulation with the grazing periodic trajectory. Here, 100 values of $q$ are sampled at local minima (in time) in the tip-sample separation after an initial transient period of 1000 cycles of the drive. This is repeated as the value of $d_0$ is decreased in steps of 0.005 nm. For the case of a periodic solution with period equal to the drive period, the data yields a single point for some particular value of $d_0$, whereas a scatter of points corresponds to more complex dynamics. For example, near $d_0 = 29.3$ nm a periodic response with period equal to three times that of the driving force is found, whereas for $d_0 = 29.6$ nm the response appears highly irregular. The figure highlights the distinct difference between the pre-grazing periodic trajectory and the post-grazing steady-state attractors. As an example, Figure 3.3 shows that the post-grazing oscillatory dynamics are characterized by a recurrent (albeit not on every close approach) occurrence of a liquid column between the tip and the sample surface.

The family of steady-state attractors transitions to a periodic steady-state attractor at $d_0 \approx 28.46$ nm. The corresponding branch of periodic trajectories may be continued using numerical continuation both for increasing and decreasing values of $d_0$ (see branch 2 in Figure 3.2 and the solid curve on the left side of the upper panel of Figure 3.3). There thus exists an interval of values of $d_0$ for which the periodic trajectories coexist with the more complicated steady-state attractors. A similar observation is again made near $d_0 \approx 2.17$ nm, where the branch of periodic trajectories again terminates at a grazing trajectory (cf. lower panel of Figure 3.3). Indeed, the family of steady-state attractors that appears around $d_0 \approx 3.3$ nm persists until $d_0 \approx 1.4$ nm where yet another branch of periodic trajectories
Figure 3.3: Steady-state attractors found using forward-time simulation. Here, 100 values of $q$ have been sampled after an initial transient period of 1000 cycles of the drive. Separate simulations are performed as the equilibrium separation $d_0$ is varied in steps of 0.005 nm. The gray solid curves on the left and right sides of the figure represent segments of branches of periodic solutions found using pseudo arclength continuation. The upper panel corresponds to the segment between points $A$ and $B$ and the lower panel corresponds to the segment between points $C$ and $D$ shown in Figure 3.2.

is found (labeled branch 3 in Figure 3.2). This branch corresponds to trajectories with $i$ identically equal to 1, i.e., for which the meniscus is maintained throughout the oscillation, and terminates at either end at points corresponding to grazing periodic trajectories.

Using forward-time simulation for a large set of initial conditions in a trial and error manner it was possible to locate and trace a second periodic steady-state attractor under variations in $d_0$ shown as the lower segment of branch 4 in Figure 3.2. As suggested by the figure,
it is not possible to continue this steady-state attractor past $d_0 \approx 28.98$ nm. Instead, initial conditions on the steady-state attractor eventually converge to the original steady-state attractor as $d_0$ is increased beyond the critical value. The fold in the branch of periodic solutions corresponds to a saddle-node bifurcation at which two branches of periodic solutions merge. In this case, the upper branch consists of unstable periodic solutions. Both branches terminate near $d_0 \approx 1.2$ nm in grazing bifurcations shortly after additional folds and changes in solution stability.

As discussed in the previous paragraphs, each branch of periodic steady-state solutions appears to terminate at a grazing trajectory that achieves grazing contact with a state-space surface on which an associated event function equals zero. Figure 3.4 shows the periodic trajectories found at each of the terminal points in Figure 3.2. Four of these, namely the trajectories labeled $A$, $B$, $C$, and $E$ achieve grazing contact with the state-space surface corresponding to the transition between $i_b = 0$ and $i_a = 1$, whereas the remaining trajectories, labeled $D$, $F$, and $G$, achieve grazing contact with the state-space surface corresponding to the transition between $i_b = 1$ and $i_a = 0$. As discussed previously, these transitions correspond to discontinuous changes in the tip-sample interaction force, an observation that can be placed on a causal footing through a rigorous analysis based on the technique of discontinuity-mappings [52,61].

While regular periodic trajectories with period identical to that of the driving exist only in certain ranges of $d_0$, it is not always easy to distinguish these from the irregular or high-periodic response found in other ranges without the sampling technique introduced here. Suppose that, instead of sampling, spectral analysis is performed to extract the complex amplitude of the frequency component of the response with frequency equal to that of the
Figure 3.4: Periodic solutions that achieve grazing contact with the $x_1 = d_0 - d_{on}$ plane ($A$, $B$, $C$, and $E$) or the $x_1 = d_0 - d_{off}$ plane ($D$, $F$, and $G$) in state space. The curves corresponding to $A$ and $B$ exhibit grazing contact with the $d_0 - d_{on}$ surface but have been rescaled in magnitude to fit on the figure with the other curves. Curves $C$-$G$ are not scaled in this way and represent the actual values.

Driving. As shown in Figure 3.5, there is an apparently continuous change in the magnitude and phase of the response calculated by this method and no distinct trace of the irregularity of the instantaneous response for $d_0 \approx 30.36$ nm and $d_0 \approx 28.46$ nm. A similar continuous transition also appears in the magnitude for small $d_0$, while some irregularity is still evident in the phase.
Figure 3.5: Variations in magnitude $\|A\|$ and phase $\arg A$ of the complex amplitude $A$ of the frequency component of the response with frequency equal to the excitation frequency under changes in $d_0$. Here, a discrete Fast Fourier Transform has been applied to the steady-state responses found using forward-time simulation and pseudo-arclength continuation sampled 512 times per period over 20 periods of the excitation.

3.4 Varying the Amplitude of Oscillation

In this section, we use parameter values similar to those in [17] which correspond to a free amplitude of oscillation of $A_0 = 21$ nm. These results form the basis of our investigations of power dissipation and basins of attraction presented in the Chapters 4 and 5. As found in previous sections for an amplitude of $A_0 = 30$ nm, these results demonstrate the same general behavior. This includes the coexistence of multiple steady-state attractors, the irregular
response separating regions of periodic response, and the existence of grazing trajectories corresponding to the end points of branches of periodic solutions. This suggests that our findings may provide general insight over a range of conditions. Figure 3.6 represents the amplitude-distance curve. The blue line is found for a simulation that begins with a large separation, initially the cantilever undergoes free oscillations represented by the horizontal blue line for normalized separations greater than one. The amplitude of oscillation goes to zero as the separation is decreased, and then retracts back to larger values for large separations. By searching around in parameter space using different initial conditions, the larger amplitude solution given by the red curve was found. Figure 3.7 represents the phase-distance curve.

Figure 3.6: (color) Numerical results using forward-time simulation to explore the dynamics of a tapping mode AFM. (left) The variation of maximum amplitude $q$ of the periodic solution with separation. (right) A close-up view showing the high-amplitude solution (red) jumps to the low-amplitude solution (blue). The amplitude of free oscillation is $A_0 = 21$ nm.

Figure 3.8 plots the minimum distance between the mass and the surface when the mass is fully extended toward the surface, given by $d_0 - q$. When plotted in this way: $d_0 - q > a_0$
Figure 3.7: (color) Numerical results using forward-time simulation to explore the dynamics of a tapping mode AFM. The variation of the phase (measured with the amplitude at the maximum) with separation. The high-amplitude solution is represented by red and the low-amplitude solution is represented by blue. The amplitude of free oscillation is $A_0 = 21$ nm.

indicates no contact with the discontinuity trigger function at $d_0 = a_0$ and consequently switching from $i_b = 1$ to $i_a = 2$; $d_0 - q = a_0$ indicates grazing contact; and $d_0 - q < a_0$ indicates penetration.

For the noncontacting oscillation, at a critical separation of $d_0 \approx 21.3$ nm the results indicate that the solution jumps from being simply periodic to high-periodic. The high periodic behavior persists until the separation is $d_0 \approx 17.1$ nm. At $d_0 \approx 17.1$ nm, grazing contact happens between the noncontacting oscillation and the discontinuity corresponding to the appearance of the capillary force.

This behavior continues until the separation is $d_0 \approx 17.1$ nm. Over the separation range
6.1nm < d_0 < 17.1nm, the solution remains periodic. However at d_0 ≈ 6.1 nm, the solution is high periodic until d_0 ≈ 0.6 nm where contact happens with the discontinuity corresponding to the onset of the repulsive force and the index becomes \( i_a = 2 \). The windows of high periodicity can be seen more clearly in Figure 3.8. On the other hand for the contacting oscillation \((d_0 - q < a_0)\), the index is \( i = 2 \) along the branch and the solution is periodic.

Figure 3.8: (color) Numerical results using forward-time simulation to explore the dynamics of a tapping mode AFM. (top) The variation of \( d_0 - q_{max} \) with separation. The dynamics are high periodic for windows 17.1nm < \( d_0 < 21.3 \)nm and 1.7nm < \( d_0 < 6.1 \)nm. (bottom) A close-up view of the high-periodic regions. For each value of the separation values from the final 200 oscillations are plotted.

Figure 3.9 shows that the solution trajectory crosses through the discontinuity trigger functions at \( d_0 - d_{off} \), and \( d_0 - d_{on} \).

The numerical continuation results for the AFM dynamics are shown in Figure 3.10 and 3.11. At the points labeled A, B, C, D, E, and F the state index switches between two values or
Figure 3.9: (color) Solution trajectories crosses through the discontinuity trigger functions at $x_1 = d_0 - d_{on}$ plane ($A$, $B$, $C$, and $E$) and $x_1 = d_0 - d_{off}$ plane ($D$, $F$, and $G$). The solutions corresponding to $A$ and $B$ are rescaled by a factor of 0.45 in state space to fit on the figure.

grazing happens. These locations are also associated with the locations that the solutions become unstable. The dotted lines show unstable solutions. The continuation technique provides detailed information about the stability of the solutions and the bifurcation points. Figure 3.12 represents the coexistence of the steady-state attractors and the branches of periodic solutions for two specific windows of separation distance. Figure 3.13 shows the variation of the real part of the largest eigenvalues at saddle node bifurcations. Where the eigenvalues cross the unit circle through +1 indicates a saddle node bifurcation.
Figure 3.10: (color) Variation of $d_0 - q_{\text{max}}$ with separation, $d_0$, using pseudo-arclength continuation. Solid blue lines represent stable solutions and the dashed red line represents an unstable solution. The periodic trajectories make grazing contact with the $x_1 = d_0 - d_{\text{on}}$ and $x_1 = d_0 - d_{\text{off}}$ planes at points A, B, C, D, E, F and G. The four different solution branches are labeled numerically.

3.5 General Features of the Bifurcation Analysis

Using both forward-time simulation and numerical continuation techniques tailored specifically for use with hybrid dynamical systems we have gained new insight in the steady-state cantilever dynamics during tapping mode atomic force microscopy. Several features of the bifurcation analysis found here are expected to generically occur in the cantilever response, although specific details will vary as to their location and extent under variations in system parameters. These include the coexistence of multiple steady-state attractors, the irregular response separating regions of periodic response, and the existence of grazing trajectories.
Figure 3.11: (color) Variation of the phase with separation, \(d_0\), using pseudo-arclength continuation. Solid blue lines represent stable solutions and the dashed red line represents an unstable solution. The periodic trajectories make grazing contact with the \(x_1 = d_0 - d_{\text{on}}\) and \(x_1 = d_0 - d_{\text{off}}\) planes at points A, B, C, D, E, F and G. The four different solution branches are labeled numerically.

corresponding to the termination points of branches of periodic solutions. Although the property of grazing is particular to the hybrid formulation, the qualitative existence of termination points as periodic trajectories reach regions in which rapid changes occur in the vector field is expected to persist in a smoothed model [62].
Figure 3.12: (color) Steady-state attractors found through forward-time simulation. Here, shown in red are 200 values of $q$ that have been sampled after an initial transient period of 2000 cycles of the excitation as the value of $d_0$ is varied in increments of 0.1 nm. The blue solid curves represent segments of branches of periodic solutions found using pseudo arclength continuation and are shown in full in Figures 3.10 and 3.11.

Figure 3.13: (color) Variation of the real part of the largest eigenvalues at saddle node bifurcations (corresponding to Figures 3.10 and 3.11). When the eigenvalues leave the unit circle through +1, the saddle node bifurcation happens. Red circles show the eigenvalues larger than +1 and blue circles show the eigenvalues smaller than +1.
Chapter 4

The Dissipated Power due to the Capillary Layer Interactions

This chapter presents the power dissipation in tapping mode AFM [63]. Understanding the force interactions between the tip of an oscillating micron scale cantilever and the surface of interest is central to dynamic atomic force microscopy (AFM) [20]. Measurements of the amplitude, frequency, and phase of the oscillating cantilever have been widely used to probe surface properties. The relative phase difference between the cantilever tip and the drive quantifies the energy dissipated by the tip-surface interactions [20,30–33,43,64]. As a result, phase contrast measurements offer the potential to probe surface properties otherwise inaccessible or difficult to determine. In experiment, the dissipated power is often measured indirectly using the harmonic approximation of Cleveland et al. [30]. In this approximation the cantilever dynamics are assumed to remain harmonic and the power dissipated by tip-
surface interactions is measured as the difference between the power supplied and the power
dissipated by elastic and viscous damping. The building of a quantitative link between
the power dissipated and the surface properties requires a detailed understanding of the
dissipation dynamics [32,33,43,65,66]. We explore the power dissipated in the presence of a
hysteretic and conservative tip-sample interaction force where the source of hysteresis is the
formation and rupture of a liquid bridge between the tip and sample.

The use of point-mass models with nonlinear, and often discontinuous, tip-surface force
interactions has been very useful in providing physical insight into the complex dynamics
tapping mode AFM [16–19,24,25]. We use the point-mass model of Zitzler et al. [17] that
includes long-range attraction, adhesion, and repulsion forces as well as the interactions of
capillary fluid layers covering both the tip and sample due to ambient humidity [42]. We
refer the reader to Ref. [17] for a detailed discussion of the model and we only provide the
essential ingredients necessary for our discussion. The equation of motion is,

\[ m\ddot{q} + \gamma \dot{q} + kq = F_0 \cos \omega t + F_{ts}(d,i), \]  \hspace{1cm} (4.1)

where \( q \) is the position of the mass \( m \), \( k \) is the spring constant, \( \gamma \) is the damping coefficient,
\( F_0 \) is the magnitude of the driving force, \( \omega \) is the drive frequency, \( d(t) = d_0 - q(t) \) is the
instantaneous tip-sample separation, \( d_0 \) is the equilibrium tip-sample separation and positive
\( q \) is in the direction towards the sample. The tip-surface force \( F_{ts}(d,i) \) depends upon both
d \( d \) and the current state \( i \) of the tip-surface force interactions. There are three possible
states: (1) long-range attraction; (2) long-range attraction and capillary interactions; and (3)
adhesion, capillary interactions, and repulsive contact with the surface.
The long-range attractive van der Waals force is modeled by $F_v = HR/6d^2$ for $d > a_0$ and as $F_v = HR/6a_0^2$ for $d \leq a_0$ where $H$ is the Hamaker constant, $R$ is the tip radius, and $a_0$ is the intermolecular distance. The repulsive contact force is found using the Derjaguin-Muller-Toporov (DMT) model as $F_r = -(4/3)E^*R^{1/2}(a_0 - d)^{3/2}$ where $E^*$ is an effective Young’s modulus of the tip and surface. The attractive capillary force is modeled by $F_c = 4\pi \gamma w R/(1 + d/h)$ for $d > a_0$ where $\gamma w$ is the surface energy of water, and $h$ is the thickness of the capillary water layer. For $d \leq a_0$ the capillary force is assumed to remain at the constant value given at $d = a_0$. As the cantilever approaches the surface the capillary layers merge when $d = d_{on}$ where it is assumed that $d_{on} = 2h$. The capillary force remains until the liquid bridge or meniscus is broken when $d = d_{off}$. Using geometrical arguments and DMT theory yields $d_{off} = V_1^{1/3} - V_2^{2/3}/(5R)$ where the volume of the meniscus $V = 4\pi Rh^2 + 4\pi h^3/3 + 2\pi r^2 h$ and $r = (3\pi \gamma w R^2/E^*)^{1/3}$ is the radius of the circular contact area [17,67].

### 4.1 An Approach to Compute the Exact Dissipated Power

The capillary force is strongly hysteretic ($d_{off} > d_{on}$) and yields the state dependence of $F_{ts}$. All of the tip-surface forces are conservative and this hysteresis is the only source of tip-surface dissipation. Any oscillation that breaks the capillary layer dissipates an equal and constant amount of energy given by,

$$E_{tip} = 4\pi \gamma w Rh \ln \left( \frac{d_{off} + h}{3h} \right), \quad (4.2)$$
where $E_{\text{tip}}$ is the area enclosed by the hysteresis loop of the force-distance curve. The power dissipated by breaking the meniscus is $P_{\text{max}} = E_{\text{tip}} f_0$ which is the maximum dissipation possible. The *exact* power dissipated for any complicated motion of the mass, for example for chaotic or quasiperiodic dynamics, is the fraction $f$ of the oscillations that break the meniscus

$$P_{\text{tip}} = f P_{\text{max}},$$

(4.3)

where $0 \leq f \leq 1$. $f = 0$ is for solutions that never break the meniscus yielding $P_{\text{tip}} = 0$, and $f = 1$ is for solutions that break the meniscus every pass near the surface yielding $P_{\text{tip}} = P_{\text{max}}$. In the following we use parameter values for a Si cantilever tip and a Si surface [17]: $k = 27.5$ N/m, quality factor $Q = 400$, the beam is driven at its resonant frequency $f_0 = 280$ kHz, a free amplitude of oscillation of $A_0 = 21$ nm, $H = 6.0 \times 10^{-20}$ J, $R = 20$ nm, $\gamma_w = 72$ mJ/m$^2$, $E^* = 66$ GPa, and $a_0 = 0.1$ nm.

The energy dissipated is directly related to the difference between $d_{\text{off}}$ and $d_{\text{on}}$ which is determined by $\gamma_w$, $R$, and $h$. As a result, $P_{\text{max}}$ does not depend upon the cantilever dynamics. Experimental measurements indicate that $h \approx 0.7$ nm corresponds to 100% relative humidity for a silicon surface [42]. Over the range $0.1 \text{ nm} \leq h \leq 0.7 \text{ nm}$ a curve fit to the exact result from the model yields $P_{\text{max}} \approx 4.5 h^{0.71}$ as shown in Figure 4.1.

The experimental system has rapidly changing tip-surface force interactions, which are not truly discontinuous. However, in order to draw physical insight from the mathematical model it is important to retain its features in the numerical simulations. We have performed simulations of Eq. (4.1) using the numerical techniques described in Ref. [19] that carefully treat the discontinuities and hysteresis in $F_{ts}$. We emphasize that artificially coarsening $F_{ts}$ in the numerical approach would lead to errors in the values of $f$ and thus an inaccurate
Figure 4.1: The variation of the maximum power dissipated $P_{\text{max}}$ with the thickness of the capillary layer $h$. The solid line is the exact result and the dashed line is a curve fit given by $P_{\text{max}} \approx 4.5h^{0.71}$.

determination of $P_{\text{tip}}$.

Using our numerical approach we compute precisely the fraction of trajectories $f$ that break the meniscus to yield the exact power dissipated by Eq. (4.3). The variation of $P_{\text{tip}}$ with $d_0$ is shown in Figure 4.2 for a capillary film thickness of $h = 0.2$ nm. Figure 4.2 illustrates the low-amplitude or attractive solution, regions (1)-(5), and the high-amplitude or repulsive solution, region (6). The low-amplitude solution does not interact with the solid surface and experiences a net attractive force whereas the high-amplitude solution interacts with the solid and experiences a net repulsive force. These coexisting solutions have been discussed in detail elsewhere [17,19]. Regions (1)-(5) form a dome which can be directly related to the cantilever dynamics. In region (1), $P_{\text{tip}} = 0$ and the capillary layer remains untouched. In region (2), the dynamics are quite complicated yet there is a simple trend in $f$ as shown in Figure 4.3.
Figure 4.2: (color) (a): The variation of $P_{\text{tip}}$ with $d_0$ for $h = 0.2$ nm. Regions (1)-(5) are the low amplitude solution, region (6) is the high amplitude solution. Blue circles are the exact value found using Eq. (4.3). Green squares and red triangles are found using Eq. (4.4). (b) A close-up view of the results in panel (a) near the plateau region.

For values of $d_0$ where the cantilever first breaks the meniscus $f$ is quite small indicating that this occurs only a small fraction of the time. Upon breaking the meniscus, an amount of energy $E_{\text{tip}}$ is dissipated. As a result, it takes several oscillations for the cantilever to return to interacting with the capillary layer upon which this cycle repeats (see Figure 4.5). This trend continues as $d_0$ is reduced and $f$ increases. Eventually a periodic solution is reached where every trajectory breaks the meniscus to yield the plateau labeled region (3) at $P_{\text{max}} = 1.45\, \mu$W indicated by the dashed horizontal line. As $d_0$ decreases further the periodic dynamics terminate to a solution where the cantilever tip remains entirely inside the fluid layer for several oscillations before breaking the meniscus, indicated by region (4). The meniscus must be broken for energy to be dissipated and, as a result, this also leads to a reduction in $P_{\text{tip}}$. As $d_0$ is decreased the fraction of oscillations remaining in the fluid layer increases. Eventually, this terminates and the cantilever exhibits a periodic solution.
Figure 4.3: (color) The fraction $f$ of trajectories that break the meniscus for different values of the capillary layer thickness: $h = 0.1\text{nm}$ (red, squares), $0.15\text{ nm}$ (black, deltas), $0.2\text{ nm}$ (blue, circles), $0.25\text{ nm}$ (cyan, right triangles), $0.5\text{ nm}$ (yellow, left triangles), and $0.6\text{ nm}$ (green, diamonds). The width of the plateau region is shown in Figure 4.6.

where the tip never leaves the water layer yielding $P_{\text{tip}} = 0$ indicated by region (5). These dynamics are general and are shown over a range of $h$ in Figure 4.2(b).

The magnitude of $P_{\text{tip}}$ in regions (1), (3), (5) can be predicted without knowledge of the cantilever dynamics whereas the dissipated power in regions (2) and (4) depend upon the dynamics. The origin and termination of the five regions also depends upon the dynamics. This is illustrated in Figure 4.3 for low-amplitude solutions at different values of $h$. The rise and fall of $f$ in regions (2) and (4), respectively, are similar over the range of capillary layer thicknesses explored. To quantify these trends we give curve fits for regions (2) and (4) for $h = 0.2\text{nm}$ as shown by the solid lines in Figure 4.4(a)-(b) which are $P_{\text{tip}} \approx -0.91 + 1.32d_0 - 0.04d_0^2$ for $17.2\text{ nm} \leq d_0 \leq 21.2\text{ nm}$ and $P_{\text{tip}} \approx 1.87d_0^{0.62}$ for $1.5\text{ nm} \leq d_0 \leq 6.2\text{ nm}$. As $h$ increases the initial interaction with the fluid layer occurs at larger $d_0$ because $d_{\text{on}} = 2h$ and
shifts the origin of the plateau of region (3) to larger values of $d_0$. For $h \approx 0.45\text{nm}$ region (3) disappears and a steady oscillating solution that breaks the meniscus every oscillation is never achieved. As a result $f < 1$ as shown for $h = 0.5$ and 0.6nm. The nonlinear variation of the width of the plateau region (3) with increasing $h$ is shown in Figure 4.6.

To illustrate the complexity of the dynamics, Figure 4.5(a) shows the cantilever velocity $\dot{q}$ as a function of position $q$ for the solution in region (2) with $f = 5/8$ of Figure 4.2(a). This is a close-up view of the cantilever trajectory as it comes closest to the surface and each pass near the surface is labeled with a number $n$ indicating the sequence in which the passes occur. The vertical dashed line represents the location of the undisturbed capillary layer. The location where the meniscus breaks is out of view to the left and every oscillation when retracting continues far past this location. Passes $n = 1, 6, 3$ do not contact the capillary layer.
Figure 4.5: (a): Velocity versus the position for a solution where $f = 5/8$, i.e. the cantilever breaks the meniscus 5 out of every 8 passes. The dashed line indicates the location of the undisturbed capillary layer. Oscillations (1,6,3) do not break the meniscus and oscillations (8,5,2,7,4) do break the meniscus. (b): $P_{\text{tip}}$ for the trajectory shown in panel (a) as a function of the $n^{\text{th}}$ pass near the surface. The circles are found using the harmonic approximation of Eq. (4.4) and the dashed line is their average value. The dash-dotted line is the exact value of $P_{\text{tip}}$.

and passes $n = 8, 5, 2, 7, 4$ enter the capillary layer and subsequently break the meniscus. In general, after breaking the meniscus the maximum value of $q$ for the very next pass is reduced. Subsequent passes have increasing values of amplitude until the water layer is contacted again. In the case shown, these eight passes repeat in sequence and represent the steady oscillating solution. Similar dynamics are found over all of region (2). Furthermore, the same trends are found in region (4). In this case the energy dissipated by an oscillation that breaks the meniscus tends to cause subsequent oscillations to remain in the capillary layer due to the small values of $d_0$. 
Figure 4.6: The variation of the width $w$ of the plateau given by region (3) in Figure 4.2(b) as a function $h$. Symbols represent results from numerical simulation and the solid line is a seventh order polynomial fit to guide the eye. The plateau region vanishes at $h \approx 0.45$ nm.

4.2 Comparison of Sinusoidal Approximation and Exact Dissipated Power

A commonly used approach in the experimental measurement of $P_{\text{tip}}$ is to assume sinusoidal steady-state response which yields, on resonance (i.e. $\omega = \omega_0$), [30]

$$P_{\text{tip}} = \frac{k A^2 \omega_0}{2Q} \left( \frac{A_0}{A} \sin \phi - 1 \right),$$  \hspace{1cm} (4.4)

where $A$ and $\phi$ are the amplitude and phase of oscillation, respectively. The nonlinearities from the contact and capillary models act for very short times and over very small portions of a cantilever oscillation. As a result, the response remain nearly sinusoidal and Eq. (4.4)
yields only minor errors. Figure 4.2(a) compares $P_{\text{tip}}$ found using Eq. (4.4) with the exact value from Eq. (4.3) for high and low amplitude solutions. The high amplitude solution, region (6), experiences the strong nonlinearity of the surface contact model and breaks the fluid meniscus at every oscillation. The resulting dissipation is exactly $P_{\text{max}}$. In this case, the strong nonlinearity of the contact model is significant and Eq. (4.4) deviates from the actual dissipation. Equation (4.4) underpredicts the dissipation and for very small values of $d_0$, where the influence of the contact repulsion is largest, it clearly leads to significant errors.

The low amplitude solution only interacts with the long range attraction and capillary forces and, in this case, Eq. (4.4) is quite accurate for the entire power dissipation-separation curve. For the plateau region, Eq. (4.4) slightly overpredicts the dissipation. It is interesting to consider further how Eq. (4.4) remains so accurate in regions (2) and (4) where many of the oscillations never actually break the meniscus layer and, as a result, do not contribute to the power dissipated. Figure 4.5(b) explores this further for the case of region (2) where $f = 5/8$ in Figure 4.2(a). The symbols are the power dissipated using Eq. (4.4) where the amplitude and phase have been calculated at each of the $n$ passes shown in Figure 4.5(b). The average of these is what would typically be measured experimentally and this value is shown by the dashed line. The dash-dotted line represents the exact value as determined from our simulations. It is clear that Eq. (4.4) predicts a finite value of dissipation even for trajectories that do not break the meniscus. The value of the predicted dissipation reflects the magnitude of the amplitude for a particular pass. For example, passes $n = 1$ and 4 have the smallest and largest values of dissipation, respectively. Overall, the average of this response leads to a fairly accurate result.
4.3 The Variation of Dissipated Power with Relative Humidity

We explore the variations of the dissipated power $P_{\text{tip}}$ in the regions where a significant change occurs in the magnitude of $P_{\text{tip}}$. Figure 4.7 shows the variation of $f$ in regions (2) and (4) with respect to $d_0$ using a smaller step size, $\Delta d_0 = 0.01$ nm. In region (2) for larger values of $d_0$, $f$ increases smoothly compared to the smaller values of $d_0$ as shown in Figure 4.7 (a) and (b). However, in region (4) for $d_0 = 5.76$ nm, there is a sudden decrease from $f = 1$ to $f = 0.93$ and for $d_0 = 1.43$ nm, there is another sudden change from $f = 0.33$ to $f = 0$ as shown in Figure 4.7 (c) and (d). These results suggest that operating the AFM cantilever closer to the surface, the dynamics of $f$ is more complicated than when far from the surface.

To investigate the influence of the relative humidity on the dynamics of the AFM, we study the steady state attractors using forward time simulation as a function of the water film thickness, $h$. Simulations have been performed for $h = 0.2$ nm, $h = 0.15$ nm and $h = 0.1$ nm. Figure 4.8 shows the branch of steady state attractors for these different values of humidity. Triangles show the simulations corresponding to $h = 0.2$ nm, squares for $h = 0.15$ nm, and circles for $h = 0.1$ nm. The simulation results suggest that by increasing the water film thickness, the branch of periodic attractors becomes wider as shown in Figure 4.6 and consequently the irregular or high-periodic dynamics becomes wider. Also, when the AFM oscillates in a less humid environment, the amplitude of oscillation corresponding to the branch of the low-amplitude solutions is larger than the amplitude of oscillation in an environment with more relative humidity. Table 4.1 shows that increasing $h$, the high-amplitude of oscillation remains about constant while the low-amplitude of solution decreases.
Figure 4.7: The variation of $f$ in regions (2) and (4) of Figure 4.3. Panels (a) and (b) are for region (2). For larger values of $d_0$, $f$ increases more smoothly compared to the behavior found for smaller values of $d_0$. Panels (c) and (d) are for region (4). For decreasing values of $d_0$ there is a sudden decrease from $f = 1$ to $f = 0.93$ and there is another sudden change from $f = 0.33$ to $f = 0$. 
Figure 4.8: (color) Branches of steady state attractors for different values of the water layer thickness $h$. Triangles (magenta) $h = 0.2$ nm, squares (blue) $h = 0.15$ nm, and circles (green) $h = 0.1$ nm. The variation of the minimum separation distance $d_0 - q_{max}$ between the cantilever and the surface is plotted versus the equilibrium separation $d_0$.

for each $d_0$.

<table>
<thead>
<tr>
<th>$h$ (nm)</th>
<th>$d_0 - q_{max}$ (nm)</th>
<th>$d_0 - q_{max}$ (um)</th>
<th>difference between the low and high amplitude solutions (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1</td>
<td>0.19</td>
<td>-0.21</td>
<td>0.40</td>
</tr>
<tr>
<td>0.15</td>
<td>0.26</td>
<td>-0.23</td>
<td>0.49</td>
</tr>
<tr>
<td>0.2</td>
<td>0.35</td>
<td>-0.24</td>
<td>0.59</td>
</tr>
</tbody>
</table>

Table 4.1: Values of $d_0 - q_{max}$ as a function of $h$. The results presented here show that increasing $h$, the high-amplitude of oscillation remains about constant while the low-amplitude of solution decreases for each $d_0$.

The difference between the resulting amplitudes is more significant for the low-amplitude branches (upper three branches in Figure 4.8) than for the high-amplitude branches.
4.4 General Insights

In conclusion, our discussion is general for any hysteretic model with conservative forces yielding steady-state oscillating solutions. The fraction of trajectories experiencing hysteresis uniquely determine the power dissipated. Given an experimentally accurate model this could be used to probe the properties and dynamics of the fluid layer [40]. Furthermore, quantitative measurements of the dissipated power may provide a solid and unambiguous imaging tool for determining the morphology as well as the compositional variations of nanometric surface samples in an ambient environment.
Chapter 5

Basins of Attraction of Steady Oscillating Solutions

The increased application of micro/nanoelectromechanical systems (MEMS/NEMS) has lead to the importance of understanding nonlinear dynamics at this scale. One potential benefit is that the sensitivity and precision of some measurements can be improved while the system is driven in nonlinear region [45,68,69].

The objective of this chapter is to quantify the basins of attraction of AFM cantilevers in tapping-mode operation with the inclusion of capillary force interactions. We are interested in determining all the possible steady state solutions and in approximating the basins of attraction of these attractors. Eventually we would like to build an understanding of how these attractors change as system parameters are varied. Of particular interest is the variation of the equilibrium separation of the cantilever from the surface during surface scanning.
5.1 Problem Overview

The deterministic character of the equation of motion requires that when a steady state attractor is found, the AFM cantilever oscillates there, independent of the extent of its basin of attraction. However, in any experiment or numerical simulation, small perturbations can influence the dynamics significantly. For AFM experiments, the perturbations can be extrinsic due to the mechanical and thermal noise [70] or intrinsic due to the finite time response of the feedback electronics [3]. For numerical simulations, small perturbations exist due to the particulars of the numerical scheme as well as round-off error. It is very important that the numerical approach used is specifically tailored for systems with rapidly changing vector fields as we discuss here. Garcia and San Paulo explored the basin of attraction in the absence of the capillary fluid layer and uncovered a complex pattern of low and high amplitude steady state periodic solutions as the equilibrium distance of the cantilever is varied [3]. We have performed a large number of forward-time simulations using event handling techniques tailored for use with discontinuous systems to quantify the basin of attraction of steady oscillating solutions with the inclusion of the capillary fluid layer [71].

5.2 The Variation of the Basin of Attraction with Equilibrium Tip Surface Separation

We explore the AFM basins of attraction by numerically integrating forward the equations of motion. We have chosen to explore precisely the system parameters given by Ref. [17]. The values of the system parameters are presented in Table 3.2 where the amplitude of free
oscillation is $A_0 = 21$ nm.

We have found three different steady oscillating solutions: a periodic solution with a low amplitude, a periodic solution with a high amplitude, and irregular or high-periodic motion as previously shown in Figure 3.6. In all cases the solution oscillates at the same frequency as the driving frequency.

Figure 5.1 shows the minimum distance $d$ between the mass and the surface for the final 200 oscillations of the cantilever. We would like to highlight that our numerical simulations did not jump from one stable solution branch to another as we changed $d_0$. This is a result of our careful use of event handling techniques which incur very little numerical perturbation to the solution. This is not necessarily the case if one uses a simple forward-time integration scheme with only an if-then type logical structure to determine the choice of vector field at some time. This is also true if one attempts to smooth the tip-surface force over spatial discontinuities. In both cases the numerical approach would introduce unnecessary errors which may be large enough to cause the solution to jump from one solution branch to another.

<table>
<thead>
<tr>
<th>Case</th>
<th>$d_0$ [nm]</th>
<th>$f_H(\phi = 0)$</th>
<th>$f_H(\phi = 4\pi/3)$</th>
</tr>
</thead>
<tbody>
<tr>
<td>(i)</td>
<td>18.6</td>
<td>0.007</td>
<td>0.018</td>
</tr>
<tr>
<td>(ii)</td>
<td>16.9</td>
<td>0.493</td>
<td>0.738</td>
</tr>
<tr>
<td>(iii)</td>
<td>8.7</td>
<td>0.879</td>
<td>0.900</td>
</tr>
</tbody>
</table>

Table 5.1: The three values of $d_0$ used to compute two-dimensional projections of the basin of attraction. $f_H$ is the fraction of the initial conditions found to yield the high-amplitude periodic solution. Only the low and high amplitude solutions were encountered and the fraction of the initial conditions yielding the low-amplitude solution is $f_L = 1 - f_H$.

We have computed two-dimensional projections of the basin of attraction for the three different equilibrium cantilever separations shown in Table 5.1. For each separation we have performed 4225 simulations over a range of initial conditions in initial displacement $q(0)$ and
Figure 5.1: (color) The variation of the closest separation distance $d$ between the cantilever and the surface with equilibrium separation $d_0$. When plotted in this way the scatter in the results indicate the presence of widows of irregular or high-periodic dynamics. The windows of complicated dynamics are $17.1 \, \text{nm} \lesssim d_0 \lesssim 21.3 \, \text{nm}$ and $1.7 \, \text{nm} \lesssim d_0 \lesssim 6.1 \, \text{nm}$. For each $d_0$ the value of $d$ for the final 200 oscillations are plotted. The upper branch (blue) is the low-amplitude solution and the lower branch (red) is the high-amplitude solution. The vertical dashed lines indicate the three separations used in calculating projections of the basin of attraction.

velocity $\dot{q}(0)$ for initial phases of $\phi = 0$ and $\phi = 4\pi / 3$. Considering the three-dimensional continuous state vector $\mathbf{x}$ given by Equation 3.2, these results correspond to two separate planes in the three-dimensional space. For the $\phi = 4\pi / 3$, the magnitude of the $f_H$ is larger at each equilibrium separation.

The basins of attraction are shown in Figure 5.2. The grid spacing in initial displacement is $\Delta q_0 = A_0 / 65 = 0.323 \, \text{nm}$, the grid spacing in initial velocity is $\Delta \dot{q}_0 = A_0 \omega_d / 65 = 1.23 \, \text{mm/s}$,
and the bottom left of each colored rectangle represents the value of the initial conditions used in the simulation. Green indicates the low-amplitude periodic solution and blue represents the high-amplitude periodic solution.

Figures 5.2 (a) - (c) represent our numerical results for initial phase of $\phi = 0$. Figure 5.2 (a) illustrates that for large separations the low-amplitude periodic solution is very stable and dominates basin of attraction projection. For the separation of $d_0 = 18.6$ nm, the low-amplitude solution is found to be high periodic as well (see the dashed vertical line labeled (i) in Figure 5.1).

Figure 5.2 (b) illustrates that for intermediate separations there is a large portion of the phase space projection dominated by the low-amplitude periodic solution. However, outside this region there is a complicated pattern of initial conditions leading to the high and low-amplitude periodic solutions indicating a significant decrease in stability. Previous studies performed by Garcia et al. [3] present a simpler dynamics outside the region dominated by the low-amplitude solution. Including the capillary force interactions, as we do here, results in a more complicated dynamics. For example, for the same region of phase space, there exist several circular regions of low-amplitude solutions separated by several regions of high-amplitude solutions compared to only one circular branch found by Garcia et al. [3].

Figure 5.3 (a) and (b) represent higher resolution simulations for two separate regions of the Figure 5.2 (b). The grid spacing in displacement and initial velocity are $\Delta q_0 = 0.161$ nm and $\Delta \dot{q}_0 = 0.615$ mm/s, respectively. Using our resolution the smoothness of the basin boundaries remains an open question. Fractal basins are found in dissipative systems and contain open sets separated by nonsmooth boundaries [44]. Figure 5.3 (b) also reveals that the basin of attraction is nonsymmetric. These investigations provide a clearer insight about
the dynamics of the AFM.

Figure 5.2 (c) yields that for small separations the stability of the low-amplitude solution is lost. The basin of attraction is now dominated by initial conditions leading to the high-amplitude solution. However, the initial conditions leading to the low-amplitude solution are quite spread out and do not appear connected at the resolution available. Again, we have studied a portion of the phase space with higher resolution. Figure 5.4 reveals that the branches of the low-amplitude solution may be connected and the dynamics is quite complicated unlike the small equilibrium separation studied by Garcia et al. [3].

Figures 5.2 (d) - (f) represent our numerical results for initial phase of $\phi = 4\pi/3$. Figure 5.2 (d) shows that the low-amplitude solution dominates basin of attraction projection the same as Figure 5.2 (a).

However, Figure 5.2 (e) illustrates that the large portion of the phase space dominated by the low-amplitude periodic solution in Figure 5.2 (b) is replaced by the high-amplitude periodic solution while outside this region shows the same pattern.

Figure 5.2 (f) yields that for this initial phase the central portion of the phase space is dominated by the high-amplitude solution as opposed to the central portion of the Figure 5.2 (e).

Because of the small amount of damping present and the resulting large quality factor, the cantilever must oscillate many times for all of the transients to decay. For our results we found that 1800 cycles of the oscillation was sufficient. As a result, the basin of attraction calculation can quickly become computationally prohibitive. If one is interested in performing many calculations, significant speedup can be achieved using an optimized fortran or C code on a parallel computer running many different initial conditions simultaneously.
Furthermore, an exciting possibility is the use of cell-to-cell mapping techniques [72–74].

5.3 Quantifying the Extrinsic Noise Due to Thermal Motion

While an AFM is scanning, the topographic features of an arbitrary sample can produce large variations in the rest tip surface separation before the steady oscillating solution is found. This is an intrinsic source of noise that is very significant in experiment. An analysis of this noise requires detailed information of the sample being explored and is not included here.

Thermal fluctuations of the AFM cantilever are an important source of extrinsic noise [70, 75, 76]. The equipartition of energy theorem states that the amount of energy contained by each available mode is $k_B T/2$. Therefore, considering the potential energy of a mass-spring-damper configuration yields

$$\frac{1}{2}k_B T = \frac{1}{2} k \delta q^2. \quad (5.1)$$

where $k_B$ is Boltzmann’s constant, $T$ is the temperature, and $\delta q$ is the average fluctuation of the displacement about its equilibrium value [3, 70]. Therefore, the average magnitude of the stochastic fluctuations in the cantilever displacement due to thermal motion are,

$$\delta q = \sqrt{\frac{k_B T}{k}}. \quad (5.2)$$
Furthermore, the average fluctuation in the magnitude of the AFM cantilever velocity is

\[ \delta \dot{q} = \delta q \omega. \] (5.3)

For the cantilever considered here at room temperature this results in position fluctuations of \( \delta q \approx 0.0123 \) nm and velocity fluctuations of \( \delta \dot{q} \approx 0.0216 \) mm/s. The thermal fluctuations are quite small with respect to the resolution of basin of attraction calculations presented here.

In conclusion, using forward-time simulations that carefully treat the dynamics of this hybrid dynamical system we find the coexistence of three steady oscillating solutions: a low-amplitude periodic solution, a high-amplitude periodic solution, and solutions with irregular dynamics. We have quantified two-dimensional projections of the basin of attraction for three different values of the equilibrium cantilever-surface separation and for two different values of phase. The basin of attraction is dominated by the low-amplitude solution at large separations and by the high-amplitude periodic solution at small separations. These careful and accurate results can be used to validate other computational schemes for computing basins of attraction such as the cell-to-cell mapping approach.
Figure 5.2: (color) Two-dimensional projections of the basin of attraction of steady oscillating solutions for the three cases listed in Table 5.1. Large separation $d_0 = 18.6$ nm (a, d), intermediate separation $d_0 = 16.9$ nm (b, e), and small separation $d_0 = 8.7$ nm (c, f). Left column shows $x(t = 0) = (q, \dot{q}, 0)$ and right column shows $x(t = 0) = (q, \dot{q}, 4\pi/3)$. Blue rectangles represent initial conditions that yield high-amplitude periodic solutions and green rectangles represent initial conditions that yield low-amplitude periodic solutions. For the separations considered here there were no steady-state solutions with irregular dynamics.
Figure 5.3: (color) Simulations with higher resolution in the grid of initial conditions for two regions shown in Figure 5.2 (b). Blue rectangles represent initial conditions that yield high-amplitude periodic solutions and green rectangles represent initial conditions that yield low-amplitude periodic solutions.

Figure 5.4: (color) Simulations with higher resolution in the grid of initial conditions to explore in more detail a portion of Figure 5.2 (c). Blue rectangles represent initial conditions that yield high-amplitude periodic solutions and green rectangles represent initial conditions that yield low-amplitude periodic solutions.
Chapter 6

Conclusions

This dissertation focused on an investigation of the dynamics of the tapping mode AFM. Since the interaction forces between the AFM cantilever tip and the sample are discontinuous, nonlinear, and hysteretic, we used numerical techniques suitable for hybrid dynamical systems. Using both forward-time simulation and pseudo-arclength continuation techniques specifically designed for hybrid dynamical systems we have studied the steady-state cantilever dynamics. The bifurcation analysis revealed the coexistence of multiple steady-state attractors, the irregular response separating regions of periodic response, and the existence of grazing trajectories corresponding to the termination points of branches of periodic solutions.

Additionally, using forward-time simulation we quantitatively studied the dissipated power by the tip of the AFM as it interacts with the capillary fluid layer by computing the fraction of the oscillations that break the meniscus.

There is no energy dissipation for the solutions that never break the meniscus. However, more complicated dynamics for which the solution trajectory interacts with the fluid layer
once and it takes several oscillations for it to return to the interacting zone results in energy dissipation. The maximum energy dissipation corresponds to the solutions that break the meniscus every oscillation. Precise measurements of the dissipated power can be used in determining the compositional variations of nanometric surface samples.

In the last section of this dissertation, we investigated the basin of attraction of the AFM. Regardless of the extent of the basin of attraction, it is expected that once the steady state solution is found, the AFM dynamics remains unchanged but small perturbations such as mechanical and thermal noise can influence the dynamics significantly. The AFM cantilever tends to stay in the state with the largest basins of attraction. Given the basins of attraction, it is possible to estimate the effect of noise on the steady state dynamics. Starting from a specific initial condition, the steady state attractor was found using forward-time simulation. This procedure is repeated for the entire phase space and the basins of attraction is generated. The results revealed that the low amplitude solution dominates the phase space for the equilibrium separations close to the free amplitude, the phase space is approximately equally divided between low and high amplitude basins of attraction for an intermediate separation, and the high amplitude solution dominates the phase space for smaller values of tip sample separations.

One possible approach to improve upon this work would be to include a more accurate model of capillary interaction forces. Theoretical investigations of the capillary force provide clear insights to the experimental studies. Macroscopic approaches such as the Zitzler model [17] and Laplace-Kelvin equation [39] investigate the capillary force and the geometric shape of the meniscus and are not the most precise approaches. In these approaches, it is often assumed that the meniscus shape can be described by two principal radii and its volume
does not change as the tip is retracted from the sample.

However, because finite molecular size effects result in large fluctuations in meniscus size and shape, these details can be overlooked in nanoscale level. Including molecular theories [77] such as molecular dynamics and Monte Carlo simulations, thermodynamic integration methods [54], would improve our model significantly.

Another important aspect of AFM which opens up some possibilities for future research is the experimental validation of our numerical and analytical techniques used to predict the behavior of the tapping mode AFM. For example, the hydrodynamic damping was assumed to be constant in our numerical simulations while it experiences significant changes at the locations of forming and breaking capillary necks due to hysteresis effects. Specifically the experiments would serve to improve our understanding of the physics of the capillary condensation and viscoelasticity of the nanometric meniscus.

The scope of this dissertation has been limited to dynamics of the AFM, but hybrid dynamical systems framework could potentially be applied to the analysis of biological systems [78]. In many biological phenomena once a specific density is met, the nature of the reaction undergoes an abrupt change. Therefore, a new set of differential equations is needed and our numerical approach designed for discontinuous systems can be a powerful tool to explain the phenomena.
Bibliography


