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Theory of amplitude modulation atomic force microscopy with and without *Q*-Control

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Abstract

The present text reviews the fundamentals of amplitude-modulation atomic force microscopy (AM-AFM), which is frequently also referred to as dynamic force microscopy, non-contact atomic force microscopy, or "tapping mode" AFM. It is intended to address two different kinds of readerships. First, due to a thorough coverage of the theory necessary to explain the basic features observed in AM-AFM, it serves theoreticians that would like to gain overview on how nanoscale cantilevers interacting with the surrounding environment can be used to characterize nanoscale features and properties of suitable sample surfaces. On the other hand, it is designed to introduce experimentalists to the physics underlying AM-AFM measurements to a degree that is not too specialized, but sufficient to allow them measuring the quantities they need with optimized imaging parameters.

More specifically, this article first covers the basics of the various driving mechanisms that are used in AFM imaging modes relying on oscillating cantilevers. From this starting point, an analytical theory of AM-AFM is developed, which also includes the effects of external resonance enhancement ("*Q*-Control"). This theory is then applied in conjunction with numerical simulations to various situations occurring while imaging in air or liquids. In particular, benefits and drawbacks of driving exactly at resonance frequency are examined as opposed to detuned driving. Finally, a new method for the continuous measurement of the tip–sample interaction force is discussed.

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1. Introduction

Since the invention of the scanning tunneling microscope (STM) in 1981 [1], a whole family of surface characterization methods (summarized as *scanning probe methods*, SPMs) has been developed, which all rely on the measurement of interactions between a sharp probe tip and a sample. Depending on the sharpness of the tip and the nature of the interaction, the interaction range is more or less localized. Scanning the tip in close proximity to the sample then allows to create a map of the strength of the specific interaction as a function of the position.

Besides the above-mentioned tunneling current, many different interactions have been exploited in SPMs, such as thermal and electrical conductivity, capacity, surface elasticity, optical properties, etc. One of the most obvious and at the same time most successful choices has been the measurement of the *force* acting between tip and sample. In early experimental setups, a sharp tip located at or near the end of a soft leaf spring (the so-called *cantilever*) profiled the sample surface in intimate and continuous contact (*contact mode*). Maps of constant tip–sample interaction force, which were usually regarded as representing the sample's "topography", were then recovered by keeping the deflection of the cantilever constant. This is achieved by means of a feedback loop that continuously adjusts the *z*-position of the sample during the scan process so that the output of the deflection sensor remains unchanged at a pre-selected value (*setpoint value*). As overwhelmingly short-range repulsive interatomic forces are responsible for contrast formation in contact mode imaging, the technique is widely referred to as *atomic force microscopy* (AFM) [2].

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Despite the success of contact mode AFM, the resolution was found to be limited in many cases (in particular for soft samples) by lateral forces acting between tip and sample. In order to avoid this effect, it has been proven to be advantageous to vibrate the cantilever in vertical direction near the sample surface. AFM imaging with oscillating cantilever is often denoted as *dynamic force microscopy* (DFM).

Similar to contact-mode AFM imaging, tip–sample forces in topographical DFM imaging are mostly short-ranged and they might be repulsive as well as attractive in nature. Since the oscillation amplitude of the oscillating cantilever are typically much higher as the interaction range of these forces, the tip "feels" the influence of the surface only during a short period of an individual oscillation, making nanoscale cantilever dynamics in atomic force microscopes inherently non-linear. To further complicate matters, cantilever dynamics is additionally governed by the specifics of how the oscillation is performed, as several distinct methods to drive the cantilever exist. For instance, you can drive the cantilever far below, close to, exactly at, or far above one of its resonance frequencies, at a fixed frequency, or at variable frequencies that are continuously adjusted depending on certain feedback parameters.

The historically oldest scheme of cantilever excitation in DFM imaging is the *external* driving of the cantilever at a *fixed excitation frequency* chosen to be exactly at or very close to the cantilever's first resonance [3–5]. For this driving mechanism, different detection schemes measuring either the change of the oscillation amplitude or the phase shift were proposed. Over the years, the *amplitude-modulation* (AM) mode, where the actual value of the oscillation amplitude is employed as a measure of the tip–sample distance, has been established as the most widely applied technique for ambient conditions and liquids.

In vacuum, the external oscillation of the cantilever has a principle disadvantage. Standard AFM cantilevers made from silicon or silicon nitride exhibit very high Q values in this environment, what makes the response of the system slow [6]. Therefore, Albrecht et al. [6] introduced in 1991 the *frequency modulation* (FM) mode, which works well for high-Q systems and consequently developed into the dominating driving scheme for DFM experiments in ultrahigh vacuum (UHV) [7–10]. In contrast to the AM mode, this approach features a so-called *self-driven* oscillator [11,12], which, in a closed-loop setup ("active feedback"), uses the cantilever deflection itself as drive signal, thus ensuring that the cantilever instantaneously adapts to changes in the resonance frequency.

Combination of these two driving mechanisms, as pioneered by Mertz et al. [13] and Anczykowski et al. [14], results in a setup that exhibits many interesting and useful properties. Most prominently, such a setup allows the active modification of the cantilever damping by the controlled increase or decrease of the apparent ("effective") Q-factor of the system. Therefore, this method has been named *Q-Control*. Its features have been be used in different ways. For example, the Q-factor can be increased to lower the maximum forces acting between tip and sample in air [14]. In similar experiments carried out in liquids, the active decrease of the damping was shown to enhance image quality [15–21]. In contrast, the possibility to actively reduce the quality factor allows to increase the scan speed in DFM experiments performed in air [22,23]. Other applications include the use in shear force microscopy [24,25], ultrasonic atomic force microscopy [26,27], or Q-controlled dynamic force spectroscopy (QC-DFS) [28].

In this review, we present a detailed theoretical analysis of the basic features of amplitude-modulation atomic force microscopy (AM-AFM) with and without active Q-Control by both analytical as well as numerical methods. For this purpose, we first highlight the similarities and differences between externally driven and self-driven cantilevers before we discuss the theoretical background of AM-AFM including Q-Control. The comparison shows how Q-Control allows to increase or decrease the cantilever damping and how the peak in the resonance curves can be shifted. Subsequently, analytical formulas describing AM-AFM imaging are developed that explicitly include tip–sample interactions. This analysis is then employed in a closer investigation of specific features of Q-controlled AM-AFM. First, we explain how Q-Control helps under ambient conditions to restrict tip–sample forces to low values well within the attractive regime, which might prevent sample damage, and how similar results might be achieved by carefully selecting the driving frequency to values slightly off the first resonance frequency. Next, we show how the tip–sample interaction potential can be recovered from the simultaneous acquisition of amplitude-vs.-distance curves and phase-vs.-distance curves. Finally, we examine the effect of Q-factor tuning on operation in liquids. It is found that Q-Control activation can lower the tip–sample indentation, what is likely to represent the origin for the often reported improved image quality of AFM micrographs recorded in liquids with active Q-Control.

2. General theory of AM-AFM

2.1. Terminology

To avoid confusion with other literature, it might be helpful to start with some words regarding the terminology used in the following. Due to the frequently occurring intermitted contact between tip and sample at the lowest point of the oscillation, the AM mode introduced above has often been denoted as "tapping mode" [4]. Over the years, use of the term "tapping mode" has then evolved into a synonym for the AM mode in many publications, disregarding whether the tip is actually making intermitted contact or not. On the other hand, if it is the operator's firm believe that no contact is established during the oscillations, the AM mode is sometimes also referred to as "non-contact" mode. Please note, however, that the term "non-contact atomic force microscopy" is more often employed in connection with the FM mode, which is mostly applied in UHV (see, e.g., [8]), than in connection with the AM mode. In this paper, we will always use the expression "amplitude modulation atomic force microscopy" (AM-AFM) to describe the driving technique, and specify separately whether intermitted contact is made or not. In some instances, however, we will

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