Tip-surface interactions in dynamic atomic force microscopy

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Abstract. In atomic force microscopy (AFM) tip-surface interactions are usually considered as functions of the tip position only, so-called force curves. However, tip-surface interactions often depend on the tip velocity and the past tip trajectory. Here, we introduce a compact and general description of these interactions appropriate to dynamic AFM where the measurement of force is restricted to a narrow frequency band. We represent the tip-surface interaction in terms of a force disk in the phase space of position and velocity. Determination of the amplitude dependence of tip-surface forces at a fixed static probe height allows for a comprehensive treatment of conservative and dissipative interactions. We illuminate the fundamental limitations of force reconstruction with narrow band dynamic AFM and we show how the amplitude dependence of the Fourier component of the force at the tip oscillation frequency, gives qualitative insight into the detailed nature of the tip-surface interaction. With minimal assumptions this amplitude dependence force spectroscopy allows for a quantitative reconstruction of the effective conservative tip-surface force as well as a position-dependent damping factor. We demonstrate this reconstruction on simulated intermodulation AFM data.

Keywords: atomic force microscopy, measurement of force, mechanical resonators, MEMS/NEMS, dissipation, intermodulation

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1. Introduction

For the understanding of surface reactions and the characterization of materials it is desirable to measure local forces close to a sample surface. The most common method to measure these surface forces is atomic force microscopy (AFM)\[1\]. Historically, the first force measurements were static measurements for which the force is presented as a scalar function of the static tip-sample separation, the so-called force curve\[2, 3\]. This representation is sufficient for conservative forces but the total tip-surface force may also contain contributions from dissipative forces. Since dissipative forces depend on probe velocity and past trajectory, dynamic force spectroscopy methods are required for their measurement. Moreover, the visualization of dissipative forces as a function of position is valid only for a specific probe trajectory and simple force curves cannot capture the full character of the interaction. Despite the development of several dynamic methods\[4, 5, 6, 7, 8, 9, 10, 11\] surface forces are still usually treated as functions of the probe position only and represented by simple force curves.

Here, we present a comprehensive framework for the representation and analysis of complex surface forces as they are measured by dynamic AFM. We concentrate on the most common modes of dynamic AFM: amplitude-modulated AFM (AM-AFM) and frequency-modulated AFM (FM-AFM), which can be considered as narrow frequency band methods\[12\]. We explore the fundamental limit of force reconstruction with narrow band dynamic AFM at fixed probe height and show how minimal assumptions allow for a quantitative reconstruction of the tip-surface interaction.

1.1. Cantilever-based dynamic force measurements

At the heart of the AFM apparatus is a micro-cantilever with a sharp tip. The cantilever is firmly clamped at one end and the tip is located at the other end which can move freely. It is assumed that surface forces only act on the tip whereas the rest of the cantilever does not experience significant surface forces. In dynamic AFM an additional external drive force is applied to maintain an oscillatory motion. Thus, the dynamics are governed by the force between tip and surface, the external drive force and the properties of the cantilever beam.

Since the cantilever is a three-dimensional continuum object its motion is usually described by the amplitudes of different oscillation eigenmodes. In general, these
modes can cause the cantilever to bend in all directions in space. However, the cantilever is positioned such that the softest flexural modes bend the beam in a plane orthogonal to the surface plane. We restrict ourselves to the case where only these flexural modes are excited by the drive force. Due to this experimental configuration the cantilever is much more susceptible to the component of the tip-surface force which is orthogonal to the surface plane. This component of the force is typically the most dominant component and the influence of lateral force components is considered negligible. In this case the cantilever acts as a mechanical projector which reacts only to one component of a three dimensional force vector field.

The deflection $w$ of a cantilever of length $L$ orthogonal to surface is described by a one dimensional Euler-Bernoulli equation\[13\]

$$EI \frac{\partial^4 w(x, t)}{\partial x^4} + \mu \frac{\partial^2 w(x, t)}{\partial t^2} = F(x, w(x), t) \tag{1}$$

where $E$ is the Young’s modulus, $I$ is the second moment of area, $\mu$ is the mass per unit length of the cantilever, $x$ is the position coordinate along the cantilever beam and $t$ is the time variable. The force term $F$ includes the surface forces acting as a point-like load at position $x = L$, the external drive force and the hydrodynamic damping due to the surrounding medium\[14\]. To express the solution to equation (1) in terms of the oscillation eigenmodes, one performs a separation of variables and obtains a solution for $w(x, t)$ that is a linear combination of different mode shapes $\Phi^{(n)}(x)$ with time-dependent amplitudes $q^{(n)}(t)$,

$$w(x, t) = \sum_{n=1}^{\infty} q^{(n)}(t) \Phi^{(n)}(x) \tag{2}$$

where the mathematically orthogonal modes $\Phi^{(n)}$ have different resonance frequencies $\omega_0^{(n)}$.

In most dynamic AFM modes only the first eigenmode is externally excited. The spectrum of the resulting tip motion is then confined in a narrow frequency band around the first flexural resonance frequency $\omega_0^{(1)}$\[12\]. Due to the fact that the resonance frequencies of higher eigenmodes are not integer multiples of the first resonance frequency $\omega_0^{(1)}$ and that the eigenmodes have high quality factors, only the first mode contributes to the cantilever motion and the tip motion $z(t) = w(x = L, t) + h$ can be approximated as

$$z(t) \approx q^{(1)}(t) \Phi^{(1)}(L) + h \tag{3}$$
where \( h \) is the static probe height above the surface. The time-dependence is given by an effective harmonic oscillator equation\(^{[15, 16]}\)

\[
\ddot{z} + \frac{\omega_0^{(1)}}{Q^{(1)}} \dot{z} + k_c^{(1)} (z - h) = F_{\text{drive}}(t) + F_{\text{ts}}(z, \dot{z}, \{z(t)\})
\]  

(4)

where the dot denotes differentiation with respect to the time \( t \), \( Q^{(1)} \) is the quality factor of the first flexural resonance, \( k_c^{(1)} \) the effective mode stiffness, \( F_{\text{drive}} \) the time-dependent external drive force and \( F_{\text{ts}} \) the force between tip and surface. Thus, equation (4) reduces the basic physics of dynamic AFM to that of a damped harmonic oscillator which is moving in an nonlinear force field \( F_{\text{ts}} \) and is subject to a time-dependent drive force \( F_{\text{drive}} \).

Simulations starting from a given tip-surface force reveal multiple oscillation states\(^{[17, 18]}\) and period multiplications\(^{[19]}\), qualitative features of the dynamics which have also been observed in experiments\(^{[20, 21, 22]}\). However, the fundamental challenge in AFM is actually the inverse problem: Given an accurate and well-calibrated measurement of the dynamics \( z(t) \), how can we quantitatively determine the tip-surface force. Different sophisticated methods have been developed to solve this inverse problem in the framework of the single harmonic oscillator model\(^{[4, 5, 6, 7, 8, 9, 10, 11]}\) but they usually assume simple forms of the tip-surface force \( F_{\text{ts}} \) in which the interaction depends on the instantaneous tip position only.

2. The force disk

2.1. General properties of surface forces

Surface forces depending on the instantaneous tip position and velocity can be represented as two dimensional function in the \( z-\dot{z} \) phase plane. The representation of a force depending on the history of the tip motion \( \{z(t)\} \), for example hysteretic forces due to capillary formation or chemical bonds, is more difficult. However, for high quality factor oscillators with large stored energy, the motion is restricted to a narrow band near resonance and the steady state motion is well approximated by a sinusoidal trajectory. For a sinusoidal drive signal at the first resonance frequency the motion is then given by\(^{[23]}\)

\[
z(t) = A \cos(\omega_0^{(1)} t + \phi) + h
\]

(5)

where \( A \) is the oscillation amplitude, \( \phi \) is the phase lag with respect to the drive. The motion orbits defined by equation (5) do not intersect each other in the \( z-\dot{z} \)
phase plane and thus every point in the phase plane can be mapped to a unique
tip trajectory. We assume that the force along each trajectory does not depend on
previous oscillation cycles. In this manner we can incorporate the dependence of the
force on the past tip trajectory into the dependence on the instantaneous tip position
and velocity.

In every experiment there exists a maximum oscillation amplitude $A_{\text{max}}$ and
velocity $v_{\text{max}}$. We normalize the tip position and velocity to the maximum amplitude
and velocity and subtract the static probe height,

$$x \equiv \frac{z - h}{A_{\text{max}}}$$

$$\dot{x} \equiv \frac{\dot{z}}{v_{\text{max}}}$$

to obtain a new force function on the closed unit disk in the $x$-$\dot{x}$ plane,

$$F_{\text{ts}}^{(x)}(x, \dot{x}) = F_{\text{ts}}(A_{\text{max}}x, v_{\text{max}}\dot{x})$$

The motion defined by equation (5) corresponds to circular orbits in the $x$-$\dot{x}$ plane
with a maximum radius of 1 as shown in fig. 1. Thus, it is sufficient to define the
force function $F_{\text{ts}}^{(x)}$ as a function on the closed unit disk in the $x$-$\dot{x}$ plane. This force
disk is a compact and general description of the interaction between the tip and the
surface in narrow band dynamic AFM.

One regularly distinguishes between conservative and non-conservative or
dissipative surface forces. The force during a complete tip oscillation cycle can readily
be decomposed into an effective conservative and an effective non-conservative force
by finding the symmetric and anti-symmetric part of the measured force around the
lower turning point of the tip motion[5, 7]. This decomposition ensures that the
energy dissipation integral

$$E_{\text{dis}} = \oint_{C[z(t)]} F_{\text{ts}} \ dz = \int_0^T F_{\text{ts}}(z(t), \dot{z}(t), \{z(t)\}, \dot{z}(t)) \ dt$$

equals zero for the effective conservative force. In contrast, dissipative forces are
responsible for the energy dissipation. One should note that forces of different
physical origin can contribute to the total surface force. Each of the effective forces
may therefore contain contributions from forces which alone would be of purely
conservative, purely dissipative or of mixed character.
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Figure 1. Force disk in the $x$-$\dot{x}$ plane of a purely conservative force. For typical values of the static probe height and the oscillation amplitude the interaction is very localized at the surface. The depicted orbits correspond to purely sinusoidal tip motion. The orbits do not intersect each other so every point in the plane can be mapped to a unique oscillation amplitude. There is one value of $F_I$ and $F_Q$ for each orbit which we assign to the lower turning point.

In the same manner we are able to decompose the force disk into an effective conservative disk $F_c^{(x)}$ and into an effective dissipative disk $F_{nc}^{(x)}$,

$$F_{ts}^{(x)}(x, \dot{x}) = F_c^{(x)}(x, \dot{x}) + F_{nc}^{(x)}(x, \dot{x}),$$

by requiring that for the conservative force disk, no energy is dissipated during one complete oscillation cycle of the sinusoidal tip motion. This implies that the conservative force disk is symmetric with respect to the x-axis whereas the non-conservative force disk is anti-symmetric:

$$F_c(x, -\dot{x}) = F_c(x, \dot{x})$$
$$F_{nc}(x, -\dot{x}) = -F_{nc}(x, \dot{x})$$
Functions on the unit disk are naturally expressed in polar coordinates so we perform a change of variables to obtain a force function $F_{ts}^{(r)}$ that depends on the normalized amplitude $r$ and the instantaneous phase of the oscillation $\theta$,

$$F_{ts}^{(x)}(x, \dot{x}) \rightarrow F_{ts}^{(r)}(r, \theta).$$

(13)

Similar to functions on the unit sphere $F_{ts}^{(r)}$ can be expanded into a set of orthogonal functions such that

$$F_{ts}^{(r)}(r, \varphi) = \sum_{n=0}^{\infty} \sum_{m=-n}^{n} a_n^{(m)} Z_n^{(m)}(r, \theta).$$

(14)

A natural choice of the basis functions $Z_n^{(m)}$ are the Zernike polynomials which can be defined as

$$Z_n^{(m)} = \begin{cases} R_n(|m|)(r) \cos(m\theta) & \text{for } m \geq 0 \\ R_n(|m|)(r) \sin(m\theta) & \text{for } m < 0 \end{cases}$$

(15)

where the polynomials $R_n(|m|)$ are given by

$$R_n(|m|)(r) = \begin{cases} (-1)^k (n - k)! \sum_{k=0}^{(n-|m|)/2} \frac{1}{k!((n+m)/2-k)!((n-m)/2-k)!} r^{n-2k} & \text{for } m - n \text{ even} \\ 0 & \text{for } m - n \text{ odd} \end{cases}$$

(16)

and fulfill the orthogonality relation

$$\int_0^1 R_n(|m|)(r) R_{n'}(|m|)(r) r \, dr = \frac{1}{2(n+1)} \delta_{n,n'}.$$  

(17)

We will use this Zernike expansion of the force disk in the following section to investigate which parts of the force disk are measurable with narrow band dynamic AFM.

2.2. Probing the force disk with dynamic narrow frequency band AFM

In dynamic AFM the tip-surface force can also be considered as a time-dependent force acting on the oscillating tip. In AM-AFM and FM-AFM only the Fourier component of the time-dependent tip-surface force at the tip oscillation frequency is measurable above the noise floor. Higher frequency components of the force are filtered out by the high-quality factor resonance of the cantilever. The force Fourier component at the oscillation frequency can be expressed as a real-valued component $F_I$ that is in-phase with a sinusoidal tip motion and a real-valued component $F_Q$
that is quadrature to the tip motion\[12\]. With equation (5) we can write $F_I$ and $F_Q$ as two integral equations

$$F_I = \frac{1}{T} \int_0^T F_{ts} \left( A \cos\left(\omega_0^1 t\right) + h, -\omega_0^1 A \sin\left(\omega_0^1 t\right) \right) \cos\left(\omega_0^1 t\right) dt \tag{18}$$

$$F_Q = \frac{1}{T} \int_0^T F_{ts} \left( A \cos\left(\omega_0^1 t\right) + h, -\omega_0^1 A \sin\left(\omega_0^1 t\right) \right) \sin\left(\omega_0^1 t\right) dt \tag{19}$$

The component $F_I$ is the so-called virial of the tip motion which is only affected by the effective conservative force\[23\]. In contrast, $F_Q$ is connected to the dissipative interaction and comparison with the energy dissipation integral in equation (9) yields

$$E_{\text{dis}} = -2\pi AF_Q. \tag{20}$$

Through their dependence on the tip motion $z(t)$ and the tip velocity $\dot{z}(t)$ the force components $F_I$ and $F_Q$ are functions of the oscillation amplitude $A$ and the static probe height $h$. Alternatively, $F_I$ and $F_Q$ can be considered as functions of $h$ and the lower turning point $z_{\text{min}}$ such that at fixed static probe height, the force disk and the $F_I(A)$ and $F_Q(A)$ curves share the same position axis as shown in figure 1.

While imaging with conventional dynamic AFM the feedback is working to keep the oscillation amplitude constant. Thus, only one value for $F_I$ and $F_Q$ is measured. Therefore, the combination of imaging and force measurement is not possible in conventional dynamic AFM and most force spectroscopy techniques rely on a measurement of the $h$ dependence of $F_I$ and $F_Q$. As an alternative approach, we recently introduced the rapid measurement of the oscillation amplitude dependence of $F_I$ and $F_Q$ at fixed static probe height with Intermodulation AFM\[12\]. To understand what information about the force disk can be extracted from a measurement of $F_I(A)$ and $F_Q(A)$ we insert the Zernike expansion of the tip-surface force disk, equation (14), into the integral equations (18) and (19)

$$F_I(A) = \int_0^{2\pi} \sum_{n=0}^{\infty} \sum_{m=0}^{n} a_n^{(m)} R_n^{(m)}\left(\frac{A}{A_{\text{max}}}\right) \cos(m\theta) \cos(\theta) d\theta$$

$$= \pi \sum_{n=0}^{\infty} a_n^{(1)} R_n^{(1)}\left(\frac{A}{A_{\text{max}}}\right), \tag{21}$$

$$F_Q(A) = \int_0^{2\pi} \sum_{n=0}^{\infty} \sum_{m=1}^{n} a_n^{(m)} R_n^{(m)}\left(\frac{A}{A_{\text{max}}}\right) \sin(m\theta) \sin(\theta) d\theta$$

$$= \pi \sum_{n=0}^{\infty} a_n^{(-1)} R_n^{(-1)}\left(\frac{A}{A_{\text{max}}}\right). \tag{22}$$
We note that $F_I(A)$ and $F_Q(A)$ only depend on the coefficients $a_n^{(\pm 1)}$ with $m = \pm 1$ where the individual coefficients can be recovered by using the orthogonality relation \[17\]. This implies that the measurable information in narrow band dynamic AFM is fundamentally limited. It is not possible to reconstruct an arbitrary force disk from the experimentally available quantities $F_I(A)$ and $F_Q(A)$. Nevertheless, the $F_I(A)$ and $F_Q(A)$ curves provide qualitative insight into the interaction between tip and surface.

The fundamental limitation at fixed static probe height is due to the limited frequency band. To increase the measurable information additional frequency bands have to be considered. One way to achieve this is to externally excite multiple cantilever eigenmodes\[26, 27, 28\]. In liquid environments higher eigenmodes exhibit measurable response even in the absence of an external drive, due to the mode’s low quality factor\[29, 30\]. However, multiple eigenmodes require a higher dimensional phase space description of the dynamics and the representation of force must be appropriately adapted. Another way to introduce additional frequency components is the measurement of higher harmonics of the tip motion, although higher harmonics are only measurable under special conditions\[6, 31\] or with specialized cantilevers\[32\].

### 2.3. Examples of force disks

In figure 2 four examples of the force disk for physically relevant model forces are shown in the $z$-$\dot{z}$ plane. Since the interaction is very localized we focus on to the region close to the surface. In figure 2a, a conservative van der Waals-Derjaguin-Muller-Toropov (vdW-DMT) force is shown which is given by equation \[A.1\]\[33\]. The symmetry of the force disk with respect to the velocity and the fact that $F_Q$ equals zero for all motion turning points, is due to the vdW-DMT force being purely conservative. From the $F_I(A)$ curve we can conclude that the conservative force can be decomposed in two regions of different character. Further away from the surface $F_I$ is positive which corresponds to an average attractive force acting on the tip. When the tip makes contact with the surface $F_I$ quickly becomes negative due to the rapid turn-on of the strongly repulsive force.

Figure 2b shows a force disk that models a long-range dissipative force\[34\]. The model builds on a conservative vdw-DMT force with a Hamaker constant whose value is different for the tip approach and retract. This discontinuous behaviour is defined by equation \[A.2\] and appears in the force disk as a discontinuous asymmetry with
respect to the velocity. The force shown in figure 2b illustrates how the addition of a so-called dissipative interaction can modify the effective conservative tip-surface force. Despite the fact that the vdw-DMT parameters are the same as for figure 2a, the $F_I$ curves differ for the two models. The added “dissipative” interaction actually contains an effective conservative component, which makes the total effective conservative force more attractive and less repulsive. The nonzero values of $F_Q$ indicate the presence of a dissipative force which is already present relatively far away from the surface. Moreover, from the shape of the $F_Q$ curve we can draw conclusions about the nature of the dissipative interaction: Further away from the surface $F_Q$ is inversely proportional to the squared oscillation amplitude. Below the contact point at $z = 0$, the derivative of $F_Q$ with respect to the oscillation amplitude reveals that $F_Q$ is inversely proportional to the oscillation amplitude in this region. This behaviour indicates that the energy dissipated during each oscillation cycle grows linearly with oscillation amplitude from the contact point on.

In contrast to figure 2b the force displayed in figure 2c is an example of an additional dissipative interaction that does not modify the effective conservative force. The force is defined in equation (A.3) and combines a vdw-DMT force with a position-dependent viscous damping\[^{35}\]. Due to the linear dependence of the force on the tip velocity the $F_I$ curve in fig 2c does not differ from the $F_I$ curve for the purely conservative vDW-DMT force in figure 2a. The damping coefficient for the force in figure 2c depends exponentially on position, which results in a dissipated energy that is proportional to the product of the oscillation amplitude and modified Bessel function of first kind in the oscillation amplitude\[^{35}\]. Hence, the $F_Q$ curve also follows a modified Bessel function of first kind.

Physically different interactions are encountered for capillary surface forces which can be due to adsorbed water layers on the surface. The common model is based on a vDW-DMT force with an additional hysteretic adhesion force that turns on and off instantaneously when the tip passes certain threshold positions as defined in equation (A.4)\[^{36}\]. Up to a threshold position $z_{on}$ the force disk is symmetric with respect to velocity. At the threshold position the tip makes contact with the water layer on the surface which results in an additional attractive force. Due to this adhesion a capillary neck builds up between the tip and the surface when the tip retracts again. This neck breaks at a position $z_{off}$ that is different from $z_{on}$. The water neck effectively extends the region of strong attraction during the tip retract. The abrupt force jumps are also visible in the $F_I(A)$ and $F_Q(A)$ curves. Since the
dissipated energy is independent of the oscillation amplitude, the $F_Q(A)$ curve is inversely proportional to the oscillation amplitude which becomes more pronounced in the derivative of $F_Q$ with respect to the oscillation amplitude.

For all example surface forces considered here the $F_I(A)$ and $F_Q(A)$ curves give insight into the character of effective conservative and dissipative interactions between the tip and the surface. With ImAFM\[37, 38\] these curves are measured at each image point while scanning at imaging speeds\[12\]. This rapid and information-rich data acquisition technique allows for an enhanced interpretation of imaging contrast, a polynomial reconstruction of the tip-surface force\[39, 40\] and the extraction of material properties\[41\].

3. Amplitude-dependence force spectroscopy (ADFS)

The ultimate goal of dynamic AFM is the combination of high-speed and high-resolution imaging with high accuracy force measurements. However, imaging is performed at static probe height $h$ above the surface and we have seen that in the case of fixed $h$ it is not possible to reconstruct a complete force disk from the available $F_I(A)$ and $F_Q(A)$ curves (see equations (21) and (22)). Often we can make physically well-motivated assumptions about the tip-surface interaction which effectively correlate the Zernike expansion coefficients of the force disk. Under these assumptions a measurement of the amplitude dependence of $F_I$ and $F_Q$ is sufficient for a quantitative reconstruction the tip-surface interaction. We call this method amplitude dependence force spectroscopy (ADFS) and in this section we show the reconstruction of the effective conservative tip-surface interaction as well as the reconstruction of a position-dependent viscous damping.

3.1. Reconstruction of conservative tip-surface interactions

In most cases it can be assumed that the effective conservative tip-surface force does not depend on tip velocity and tip motion history but is rather a function of the tip position only,

$$F_c(z, \dot{z}) = F_c(z).$$

$$\Rightarrow F_c^{(x)}(x, \dot{x}) = F_c^{(x)}(x)$$

We recently demonstrated how this assumption allows for the reconstruction of the effective conservative surface force $F_c$ with ADFS\[42\]. Using the equations (24) and
we rewrite the integral equation (18) for $F_I$

\[
F_I(A) = \frac{1}{2\pi} \int_0^{2\pi} F_c(A \cos \theta + h) \cos(\theta) d\theta \tag{25}
\]

\[
z = A \cos \theta \quad \frac{1}{\pi} \int_{-A}^{A} F_c(z + h) \frac{z/A}{\sqrt{A^2 - z^2}} dz. \tag{26}
\]

Usually, the interaction length of the force is small (several nm) compared to the oscillation range (tens of nm). So we can neglect the upper half of the integration interval,

\[
F_I(A) \approx \int_{-A}^{0} F_c(z + h) \frac{z/A}{\sqrt{A^2 - z^2}} dz \tag{27}
\]

\[
u \equiv z^2 = \frac{1}{2\pi A} \int_{0}^{A^2} F_c(-\sqrt{u} + h) \frac{\sqrt{u}}{\sqrt{A^2 - u}} du \tag{28}
\]

With the definitions

\[
\tilde{A} \equiv A^2 \tag{29}
\]

\[
\tilde{F}_c(u) \equiv F_c(-\sqrt{u} + h) \tag{30}
\]

\[
\tilde{F}_I(\tilde{A}) \equiv -2\pi \sqrt{\tilde{A}} F_I(\sqrt{\tilde{A}}) \tag{31}
\]

equation (28) becomes

\[
\tilde{F}_I(\tilde{A}) = \int_{0}^{\tilde{A}} \frac{\tilde{F}_c(u)}{\sqrt{\tilde{A} - u}} du. \tag{32}
\]

We note that $\tilde{F}_I(\tilde{A})$ in equation (32) is the Abel transform of the force $\tilde{F}_c(u)$. The Abel transform has a unique inverse\cite{43, 44} with which we solve equation (32) for the force $F_c(-z + h)$

\[
\tilde{F}_c(u) = \frac{1}{\pi} \frac{d}{du} \int_{0}^{u} \frac{\tilde{F}_I(\tilde{A})}{\sqrt{u - \tilde{A}}} d\tilde{A} \tag{33}
\]

\[
\Rightarrow F_c(-z + h) = -\frac{1}{z} \frac{d}{dz} \int_{0}^{z^2} \frac{\sqrt{\tilde{A}} F_I(\sqrt{\tilde{A}})}{\sqrt{z^2 - \tilde{A}}} d\tilde{A}. \tag{34}
\]

In practice, the integral in equation (34) has to be evaluated numerically which is made difficult by the square root singularity at the upper integration limit. It is
therefore advantageous to perform the substitution \( y^2 = z^2 - \tilde{A} \) in the integral to remove the singularity and to improve the numerical stability,

\[
F_c(-z) \equiv z^2 - A - \frac{2}{z} \frac{d}{dz} \int_z^\infty \sqrt{z^2 - y^2} F_I \left( \sqrt{z^2 - y^2} \right) dy.
\]

The resulting integral is then suitable for standard numerical integration methods like simple trapezoidal integration.

### 3.2. Reconstruction of position-dependent damping

Often dissipation is modeled as a position-dependent damping or friction coefficient \( \lambda(z) \) such that the dissipative force is given by

\[
F_{nc}(z, \dot{z}) = \lambda(z) \dot{z}.
\]

In this case we can reconstruct the effective damping function \( \lambda(z) \) from the amplitude-dependence of \( F_Q(A) \). We start with rewriting the integral equation (19) by using the equations (12) and (36).

\[
F_Q(A) = -\frac{\omega A}{2\pi} \int_0^{2\pi} \lambda(A \cos \theta + h) \sin^2 \theta \, d\theta
\]

\[
\equiv A \cos \theta - \frac{\omega A}{\pi} \int_A^{-A} \lambda(z + h) \sqrt{A^2 - z^2} \, dz
\]

As for the conservative force we assume that the oscillation range is bigger than the interaction range of the dissipative force such that \( \lambda(z) = 0 \) for \( z \geq 0 \),

\[
F_{Q}(A) \approx -\frac{\omega}{\pi A} \int_A^{-A} \lambda(z + h) \sqrt{A^2 - z^2} \, dz
\]

\[
\equiv A^2 - \frac{\omega}{2\pi A} \int_0^{A^2} \frac{\lambda(-\sqrt{u} + h)}{\sqrt{u}} \sqrt{A^2 - u} \, du
\]

We use definition (29) and define additionally

\[
\tilde{F}_Q(\tilde{A}) \equiv -\frac{2\pi \sqrt{A}}{\omega} F_Q \left( \sqrt{\tilde{A}} \right)
\]

\[
\tilde{\lambda}(u) \equiv \frac{\lambda(-\sqrt{u} + h)}{\sqrt{u}}
\]

to arrive at

\[
\tilde{F}_Q(\tilde{A}) = \int_{\tilde{A}}^{A} \tilde{\lambda}(u) \sqrt{\tilde{A} - u} \, du.
\]
The integral (43) represents a convolution of $\tilde{\lambda}(u)$ with $\sqrt{u}$ which can readily be solved in Laplace space (see Appendix B). For tip motion as defined by equation (3) the position-dependent damping coefficient is then given by

$$\tilde{\lambda}(\tilde{A}) = \frac{2}{\pi d A^2} \int_0^{\tilde{A}} \frac{\tilde{F}_Q(u)}{\sqrt{\tilde{A} - u}} \, du$$

$\Rightarrow \lambda\left(-\sqrt{\tilde{A}} + h\right) = -\frac{4\sqrt{\tilde{A}}}{\omega_{0,1} d A^2} \int_0^{\tilde{A}} \frac{\sqrt{u F}_Q(\sqrt{u})}{\sqrt{\tilde{A} - u}}$ (45)

3.3. Numerical results

To validate the reconstruction of the effective conservative tip-surface force and the position dependent-damping we simulate the tip motion in a scalar force field with a vdW-DMT component and exponential damping as defined by equation (A.3). To integrate the equation of motion (4) we use the CVODE solver with adaptive step-size and discrete event detection[45] to account for the piecewise definition of the tip-surface force in equation (A.3). We assume a cantilever typically used for experiments under ambient conditions with a quality factor of $Q^{(1)} = 400.0$, a stiffness of $k^{(1)} = 40.0 \text{ N/m}$ and a resonance frequency of $f_{0}^{(1)} = 300.0 \text{ kHz}$. To probe the tip-surface interaction we use an ImAFM drive scheme such that the free oscillation amplitude is modulated between 0 and 35 nm in a time window of $T = 2 \text{ ms}$ at a fixed static probe height of $h = 22.0 \text{ nm}$ above the sample surface as shown in figure 3a.

From the tip motion close to the surface the $F_I(A)$ and $F_Q(A)$ curves are obtained[12] which are then used for the numerical evaluation of the equations (34) and (45). The results are shown in figure 3. Both the reconstructed effective conservative force and the position-dependent damping are in excellent agreement with the force and damping actually used in the simulation. For the conservative force the actual and the reconstructed force deviate by at most 4.5% of the maximum force. This deviation is the result of the rapid change in the force around the contact point, which generate Fourier components with amplitudes typically below the noise level in AFM systems and these components have been neglected in the reconstruction.

We note that the interaction is localized in the region between -3.2 nm and 7.5 nm. A more optimized drive scheme would therefore modulate the oscillation amplitude only in this interaction region, thereby sampling the interaction more
smoothly since more turning points lie in the interaction region during the same measurement time\cite{46, 47}.

4. Conclusions

We have introduced a comprehensive framework for the representation of tip-surface interaction at fixed static probe height in terms of a “force disk”. This framework allows for a complete description of forces depending on the instantaneous tip position and velocity, appropriate to narrow band dynamic AFM. We show that it not possible to fully reconstruct arbitrary tip-surface interactions from narrow band dynamic AFM measurements. However, by considering different tip oscillation amplitudes, the experimental curves $F_I(A)$ and $F_Q(A)$ give insight in the character of the effective conservative and the effective dissipative interaction. With minimal additional assumptions these curves can be used for quantitative reconstructions of the effective conservative tip-surface force and the effective position-dependent damping.

The framework introduced here gives a more solid theoretical foundation for the visualization and analysis of tip-surface forces than traditional force curves. Furthermore, we expect that the notion of a force disk will inspire new multifrequency force probing schemes expanding the capabilities of conventional narrow band dynamic AFM. On the experimental side amplitude-dependence force spectroscopy enables rapid force measurements while scanning sample surfaces at normal imaging speeds. From the obtained force volume data sets different mechanical properties of the sample surface can be derived without the constraints and limitations of oversimplified interaction models.

Since the underlying physics of the theory presented here is the classical harmonic oscillator moving in an external force field. We see wide-spread applications of this method of analysis in other fields using resonant detection techniques based on different kinds of high-quality factor resonators\cite{46, 47}.

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Appendix A. Definition of model surface forces

In section 2.3 we introduced various force models of which the mathematical definitions are given here. Also given are the values of the parameters used when plotting or simulating with these models.

The basis of all models forms a vdW-DMT force which is defined as

\[
F_{\text{DMT}}(z) = \begin{cases} 
-HR/6(a_0 - z)^2 & \text{for } z \geq 0 \\
-HR/6a_0^2 + 4E^*/3R\sqrt[3]{R(-z)^3/2} & \text{for } z < 0 
\end{cases}
\]  

(A.1)

where \( H = 3.28 \cdot 10^{-17} \) J is the Hamaker constant, \( R = 10.0 \) nm is the tip radius, \( a_0 = 2.7 \) nm is the intermolecular distance and \( E^* = 1.5 \) GPa is the effective stiffness of the tip-surface system.

The long-range dissipative force is defined as

\[
F_{\text{long}}(z, \dot{z}) = \begin{cases} 
-H_{\text{app}}R/6(a_0 - z)^2 - \gamma_0 \exp(-z/z_\gamma) \dot{z} & \text{for } z \geq 0 \text{ and } \dot{z} \leq 0 \\
-H_{\text{ret}}R/6a_0^2 + 4E^*/3R\sqrt[3]{R(-z)^3/2} - \gamma_0 \exp(-z/z_\gamma) \dot{z} & \text{for } z < 0 \text{ and } \dot{z} \leq 0 \\
-H_{\text{ret}}R/6a_0^2 + 4E^*/3R\sqrt[3]{R(-z)^3/2} & \text{for } z \geq 0 \text{ and } \dot{z} > 0 \\
-H_{\text{app}}R/6(a_0 - z)^2 + \gamma_0 \exp(-z/z_\gamma) \dot{z} & \text{for } z < 0 \text{ and } \dot{z} > 0 
\end{cases}
\]  

(A.2)

which extends the vdW-DMT model by a Hamaker constant that depends on if the tip is approaching the surface \( (H_{\text{app}} = H) \) or is retracting from it \( (H_{\text{ret}} = 2H) \).

The vdW-DMT with an additional viscous damping term depending exponentially on position is given by

\[
F_{\text{exp}}(z, \dot{z}) = \begin{cases} 
-HR/6(a_0 - z)^2 - \gamma_0 \exp(-z/z_\gamma) \dot{z} & \text{for } z \geq 0 \\
-HR/6a_0^2 + 4E^*/3R\sqrt[3]{R(-z)^3/2} - \gamma_0 \exp(-z/z_\gamma) \dot{z} & \text{for } z < 0 
\end{cases}
\]  

(A.3)

where \( \gamma_0 = 3.5 \) Js is the damping factor and \( z_\gamma = 1.5 \) nm is the damping decay length.
Capillary interactions are modeled as

\[
F_{\text{cap}}(z, \{z(t)\}) = \begin{cases}
-HR & \text{for } z \geq z_{\text{off}} \\
-HR & \text{for } z \geq z_{\text{on}} \text{ and } z < z_{\text{off}} \text{ and } \zeta < z_{\text{on}} \text{ and } z \geq 0 \\
-HR & \text{for } z < z_{\text{on}} \text{ and } z \geq 0 \\
-HR & \text{for } z < z_{\text{on}} \text{ and } z < z_{\text{off}} \text{ and } \zeta < z_{\text{on}} \text{ and } z \geq 0
\end{cases}
\]

where \( \gamma_{\text{H}_2\text{O}} = 72 \cdot 10^{-3} \text{ J/m}^2 \) is the surface energy of water and \( h_{\text{H}_2\text{O}} = 0.1 \text{ nm} \) is the effective thickness of the adsorbed water layer. The state variable \( m \) is set to 1 when the tip makes contact with the water layer at \( z_{\text{on}} = 0.2 \text{ nm} \) and is set to 0 when the water neck breaks at \( z_{\text{off}} = 1.94 \text{ nm} \). In contrast to the model introduced in \[36\] we assume that the interface between the surface and the water layer is located at \( z = 0 \text{ nm} \).

**Appendix B. Inversion of the damping integral**

The reconstruction of the position-dependent damping \( \lambda(z) \) from \( \tilde{F}_Q(\tilde{A}) \) requires in the inversion of the integral equation

\[
\tilde{F}_Q(\tilde{A}) = \int_0^{\tilde{A}} \tilde{\lambda}(u) \sqrt{\tilde{A} - u} \, du \quad \text{(B.1)}
\]

We study this convolution in Laplace space where it becomes

\[
\mathcal{L}\{\tilde{F}_Q(\tilde{A})\} = \mathcal{L}\left\{\int_0^{\tilde{A}} \tilde{\lambda}(u) \sqrt{\tilde{A} - u} \, du \right\} = \mathcal{L}\{\tilde{\lambda}(\tilde{A})\} \mathcal{L}\{\sqrt{\tilde{A}}\}. \quad \text{(B.2)}
\]

The Laplace transform of the square root is \( \mathcal{L}\{\sqrt{\tilde{A}}\} = \frac{\sqrt{\pi}}{2} s^{-3/2} \) which yields

\[
\frac{1}{s^2} \mathcal{L}\{\tilde{\lambda}(\tilde{A})\} = \frac{2}{\pi} s^{-1/2} \mathcal{L}\{\tilde{F}_Q(\tilde{A})\} \quad \text{(B.3)}
\]

\[
= \frac{2}{\pi} \mathcal{L}\left\{\frac{1}{\sqrt{\tilde{A}}}\right\} \mathcal{L}\{\tilde{F}_Q(\tilde{A})\} \quad \text{(B.4)}
\]
\[
= \frac{2}{\pi} \mathcal{L} \left\{ \int_0^\tilde{A} \frac{\tilde{F}_Q(u)}{\sqrt{\tilde{A} - u}} \, du \right\} \tag{B.5}
\]

Now, we use the relation between integration in the time domain and its Laplace transform and obtain
\[
\int_0^{\tilde{A}_1} d\tilde{A}_1 \int_0^{\tilde{A}_2} d\tilde{A}_2 \tilde{\lambda}(\tilde{A}_2) = \frac{2}{\pi} \int_0^{\tilde{A}} \frac{\tilde{F}_Q(u)}{\sqrt{A - u}} \, du \tag{B.6}
\]
which we can solve for the damping function \( \tilde{\lambda}(\tilde{A}) \),
\[
\tilde{\lambda}(\tilde{A}) = \frac{2}{\pi} \frac{d^2}{dA^2} \int_0^{\tilde{A}} \frac{\tilde{F}_Q(u)}{\sqrt{\tilde{A} - u}} \, du \tag{B.7}
\]

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Figure 2. Model force disks in the $z$-$\dot{z}$ plane at a static probe height of $h = 22$ nm and a maximum oscillation amplitude $A_{\text{max}} = 25$ nm. The models all build on a purely conservative vdW-DMT force (a) and add long range adhesion(b), position-dependent viscous damping(c) and capillary interactions(d) respectively. We concentrate on the region close to the surface and display the disk sections together with the corresponding $F_I$ and $F_Q$ curves as functions of the lower motion turning point.
Figure 3. Simulated tip motion in the time domain (a). On a fast time scale the tip motion is nearly purely sinusoidal with an amplitude that is modulated on a much slower time scale. The ADFS reconstructions of the effective conservative force curve (a) and the position-dependent damping curve (b) are in excellent agreement with the reference curves used in the simulation.