Interaction sensing in dynamic force microscopy

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Abstract. A first-order perturbative theory of the motion of a harmonic oscillator interacting with a weak arbitrary force field is presented, as it pertains to dynamic force microscopy. In essence the theory corresponds to a Born approximation for the scattering of standing waves trapped in a perturbed parabolic potential. In particular, it is shown that the scattering amplitudes are related to corresponding moments, involving Chebyshev polynomials and associated metrics of the conservative interaction force, and of a generalized friction coefficient accounting for irreversible interactions. Implications for dynamic force microscopy are discussed.

1. Introduction

Dynamic force microscopy has proved to be a powerful tool for atomic scale imaging [1]–[3]. Here, the force sensor is operated in an oscillator circuit, and the interaction is sensed via induced shifts of the oscillator frequency. In general, a vibration amplitude larger than or comparable to the range of interaction is used. Consequently, the interaction is probed nonlocally. From an experimentalist’s point of view, one would like to determine the laws of interaction from measured dynamic data. Conventionally, the problem has been addressed from the perspective of relating frequency shifts to interaction potentials [4]–[7], or alternatively, by investigating the properties of the resonance curve [8]. Here, we approach the problem from a scattering point of view. In the spirit of a first-order Born approximation, the higher-order Fourier components of the tip motion, which correspond to scattered waves, are calculated for arbitrary reversible and dissipative interactions. In particular, it is shown that the amplitudes of the scattered waves can be used to reconstruct the interaction force in a direct way. Dissipative interactions have so far been discussed only rarely in the literature, although they constitute an important complementary observable [3]–[10]. In this paper, a well-defined methodology is presented that links experimental observables associated with dissipative interactions to a generalized friction coefficient.
2. Conservative interactions

2.1. General theory

Let us consider a force sensor that is vibrating at its resonance frequency. We first restrict ourselves to nondissipative, reversible interactions. Correspondingly, the tip orbital must be an even function with respect to time reversal and can thus be written as a Fourier series involving only cosine functions

\[ \psi(t) = \sum_{n=0}^\infty a_n \cos(n\omega t) \]  

(1)

where \( a_1 \) corresponds to the unperturbed vibration amplitude. For small vibration amplitudes, the dynamics of the force sensor can be described in terms of a simple harmonic oscillator with a resonance frequency

\[ \omega = \omega_0 \sqrt{1 + \frac{C_i}{C}} \]  

(2)

where \( \omega_0 \) is the resonance frequency of the free sensor of stiffness \( C \), and \( C_i \) denotes the local interaction force gradient at the tip position. To solve the nonlocal problem of an arbitrary vibration amplitude, we invoke a least-action principle

\[ \delta S = \int_0^T \left[ \omega_0^2 \psi(t) + \ddot{\psi}(t) - \frac{\omega_0^2}{C} F_{\text{int}}(\psi(t)) \right] \delta \psi \, dt \equiv 0. \]  

(3)

This yields the following set of coupled equations linking the Fourier coefficients to the interaction force [6]:

\[ a_n(1 + \delta_{n,0})\pi(\omega_0^2 - n^2\omega^2) - \frac{\omega_0^2}{C} \int_0^T F_{\text{int}} \left( z_0 + \sum_{k=0}^\infty a_k \cos(k\omega t) \right) \cos(n\omega t) \omega \, dt = 0. \]  

(4)

Here, \( z_0 \) is an experimental offset parameter fixing the position of the force sensor with respect to the sample (see figure 1(a)); \( T \) denotes the oscillation period, and \( \delta_{n,0} \) is the Kronecker symbol.

To proceed, we assume that the interaction is weak in the sense that all Fourier coefficients with \( n \neq 1 \) are small compared with \( a_1 \), which justifies the use of perturbative methods. In a first-order approximation we may therefore set \( a_k = 0 \) for all \( k \neq 1 \) in the orbital function of the integrand. Projecting the circular variable \( \omega t \) onto the Cartesian tip–sample axis, \( u = \cos(\omega t) \) (see figure 1(b)), yields

\[ a_n = \frac{2}{(1 + \delta_{n,0})\pi} \frac{\omega_0^2}{C(\omega_0^2 - n^2\omega^2)} \int_{-1}^1 F_{\text{int}}(z_0 + a_1 u)T_n(u) \frac{du}{(1 - u^2)^{1/2}} \]  

(5)

where \( T_n(u) = \cos(n \cos^{-1}(u)) \) denotes the \( n \)th-order Chebyshev polynomial of the first kind [11]. These polynomials represent the projected circular harmonics exactly like the Legendre polynomials represent the projected spherical functions. The Chebyshev polynomials form a complete orthogonal set with respect to the metric \((1 - u^2)^{-1/2}\), which weights the integrand in proportion to the relative fraction of time the tip spends at a given position. The first few polynomials are \( T_0 = 1, T_1 = u, T_2 = 2u^2 - 1, T_3 = 4u^3 - 3u \), and the higher-order polynomials are most easily obtained from the recursion relation \( T_{n+1}(u) = 2uT_n(u) - T_{n-1}(u) \).
Figure 1. (a) Schematic drawing of dynamic force sensing in which the tip performs an oscillatory motion $ψ(t)$ perpendicular to the sample surface. $z_0$: equilibrium position, $a_1$: amplitude of oscillation, $z = z_0 - a_1$: distance of closest approach. (b) Actual tip orbital versus nominal tip orbital. The latter is defined as the fundamental harmonic motion $z_0 + a_1 u$, where $u = \cos(\omega t)$ denotes the projection of the circular phase variable $\omega t$ onto the tip–sample axis. Note that as a result of the nonharmonic interaction force, the actual tip orbital deviates slightly from a pure cosine function by an amount $ΔΨ$.

The perturbation of the actual tip orbital from a pure cosine function (see figure 1(b))

$$Δψ = ψ - (z_0 + a_1 u) = a_0 + ∑_2^∞ a_n T_n(u)$$

(6)

can also be expressed in terms of Chebyshev functions. Equation (6) provides a direct link between the experimental orbital parameters, namely the tip offset $z_0$ and the vibration amplitude $a_1$, and the true motion of the tip. It will be shown below that in general, the largest correction term is a constant offset $a_0$ describing the deflection of the spring due to the average force seen by the tip.

The dynamics of the system are completely described by equation (5). The frequency shift, which by virtue of equation (2) is expressed in terms of an effective force gradient $C'_i^{\text{eff}}$, follows
from the equation for the principal harmonic amplitude $a_1$:

$$C_{\text{eff}}^i(z_0) = -\frac{2}{\pi a_1} \int_{-1}^{1} F_{\text{int}}(z_0 + a_1 u) u \frac{du}{(1 - u^2)^{1/2}}. \tag{7}$$

The frequency shift can be interpreted in simple physical terms. It reflects a weighted dipolar moment of the interaction probed by the tip. Similarly, the amplitudes of the harmonics of the tip orbital are coupled to corresponding weighted moments of the interaction force. Because of the orthogonality of these moments, one can readily reconstruct the force field in the tip–sample distance interval probed by the tip from a measurement of the amplitudes $a_n$. To this end, we express the tip–sample force in terms of a Chebyshev series

$$F_{\text{int}}(z_0 + a_1 u) = \sum_{n=0}^{\infty} f_n(z_0, a_1) T_n(u) \tag{8}$$

with

$$f_n(z_0, a_1) = \frac{2}{(1 + \delta_{n,0})\pi} \int_{-1}^{1} F_{\text{int}}(z_0 + a_1 u) T_n(u) \frac{du}{(1 - u^2)^{1/2}}. \tag{9}$$

Combining equations (5) and (9) yields the following correspondence between the expansion coefficients and the Fourier components of the tip orbital:

$$f_n(z_0, a_1) = a_n(z_0, a_1) C \left( 1 - \frac{n^2 \omega^2}{\omega_0^2} \right) = a_n(z_0, a_1)(C - n^2(C + C_{\text{eff}}^i)). \tag{10}$$

2.2. Discussion

The expansion coefficients $f_n$ define the reciprocal space to the tip–sample axis. Analogous to Fourier expansions, a large number of Chebyshev coefficients are required to represent point-like short-range forces. This situation is typical for large-amplitude dynamic force sensing in which the interaction is probed only at the very extreme of the tip oscillation at closest sample approach. In fact, it has been shown [6] that in these cases the low-order harmonic amplitudes are in essence proportional to the effective force gradient, and hence provide virtually no additional information. As a result, one would have to determine the amplitudes of a large number of high-order harmonics with great precision to obtain a satisfactory representation of the interaction force. This in turn is a difficult task as the corresponding Fourier amplitudes scale as $1/n^2$. For large vibration amplitudes it is therefore more practical to measure the effective force gradient as a function of the offset parameter $z_0$, and to subsequently recover the force law by inverting the integral equation (7) as described in detail in references [6] and [12].

On the other hand, the Chebyshev series converges rapidly if the oscillation amplitude is of the same order of magnitude as the range of interaction, which corresponds to the preferred operating mode [13]. Correspondingly, a good representation of the force law is already obtained from a few expansion coefficients. This opens up new perspectives for force microscopy. Monitoring the amplitudes of the higher harmonics enables the instantaneous reconstruction of the force field being probed by the tip. This additional information can be exploited to distinguish between chemically different species at surfaces and to implement stable feedback
Figure 2. (a) Chebyshev expansion coefficients $f_n(a_1, z_0)$ of a Morse-type force law (equation (11)) as a function of the distance of closest approach for a vibration amplitude $a_1 = 1.5\ell$, where $\ell$ denotes the range of interaction. (b) Interaction force (equation (11)), solid curve, versus tip–sample distance. The interaction can be expressed in the interval, $z_0 - a_1 \ldots z_0 + a_1$, probed by the vibrating tip in terms of a series $F_{\text{int}}(x) = \sum_{n=0}^{N} f_n(a_1, z_0) T_n((x - z_0)/a_1)$, where $T_n$ denotes the $n$th-order Chebyshev polynomial of the first kind. The expansion coefficients $f_n$ are proportional to the Fourier amplitudes of the tip orbital. Note that for a vibration amplitude comparable to the range of interaction, in this example $a_1 = 1.5\ell$, the interaction is fairly accurately represented using just a few terms of the expansion. Dotted curve: $N = 1$, dash-dotted: $N = 2$, and dashed curve: $N = 3$. (c) Orbital correction $\Delta\Psi$ (solid curve) as a function of nominal tip position. The dominant term in the correction is due to a constant deflection of the spring reflecting the average force seen by the tip ($a_0 T_0$, dashed curve). The higher-order harmonics ($a_2T_2$ dash-dotted, and $a_3T_3$ dotted curve) rapidly decrease in amplitude with increasing order.

For the sake of the argument we consider a Morse-type force law (see figure 2(b), solid curve):

$$F_{\text{int}}(x) = F \left( \exp(-2x/\ell) - \exp(-x/\ell) \right).$$  

(11)
Figure 3. Periodic tip orbital $\psi(t) = \psi(t + T)$ in the presence of dissipative interactions. Note that the time reversal symmetry is broken. For weak interactions one can define a path length $s$ using $ds = |d\psi/dt|dt$, which is conserved and which allows the interactions to be classified with respect to their symmetry properties for path inversion with respect to an extremum $s_0$.

The corresponding expansion coefficients $f_n(z_0, a_1 = 1.5\ell)$ are plotted in figure 2(a) as a function of the nominal distance of closest approach $z = z_0 - a_1$ (see figure 1(a)). Note that in an experiment, $f_0 = a_0C$ is obtained from a static measurement of the lever deflection, $f_1 = a_1C_{\text{eff}}$ is derived from the resonance frequency shift, and $f_2 = -a_2(3C + 4C_{\text{eff}})$ and $f_3 = -a_3(8C + 9C_{\text{eff}})$ are determined by measuring the amplitudes of the corresponding harmonics of the tip orbital, i.e. by means of lock-in techniques. The interaction force can be reconstructed fairly accurately using just four expansion coefficients, as shown in figure 2(b) for $z_0 = a_1$. In particular, the position of the adhesion maximum, which is an important characteristic of the interaction, can be readily determined in a single-shot experiment rather than by measuring a frequency shift curve as a function of tip displacement. Dynamic force microscopy could benefit substantially if the imaging control parameter is derived from a direct measurement of the position of the adhesion maximum rather than simply from a shift of the resonance frequency, as this would improve the reproducibility of operating conditions and thus the ease of operation.

Finally, the orbital correction is inspected. The graphs shown in figure 2(c) correspond to $z_0 = a_1$ and $a_1 = 1.5\ell$. Note that the orders of magnitude of the expansion coefficients $f_n$ are about equal (see figure 2(a)), but that, according to equation (10), the corresponding amplitudes $a_n$ scale as $f_n/(Cn^2)$. Hence, they rapidly become smaller with increasing order. For most practical purposes it is thus sufficient to restrict oneself to the lowest one or two correction terms $a_0$ and $a_2T_2(u)$ when assessing the degree of orbital distortion, which in addition scales inversely with the spring constant of the force sensor.
3. Dissipative interactions

3.1. General theory

The formalism for conservative forces can be readily extended to include dissipative forces. Let us consider periodic steady states only, that is \( \psi(t+T) = \psi(t) \), meaning that an appropriate driving force is applied to maintain an overall constant energy over one oscillation cycle. As dissipative interactions are involved, time reversal is no longer a good symmetry of the system. We assume again that the dissipative interactions are weak in the sense that the orbital function \( \psi(t) \) has exactly two local extrema which are associated with the classical turning points. Otherwise, the orbital can have any arbitrary shape.

To proceed we exploit the fact that for weak interactions the total path length of the orbital \( \int ds = L \) is conserved, where \( ds = |d\psi/dt|dt \) (see figure 3). This allows us to decompose the tip orbital into two distinct, well-defined phases corresponding to the approach, \( d\psi/ds = -1 \), and the retraction, \( d\psi/ds = +1 \), of the tip towards and from the sample, respectively. Furthermore, the orbital function is fully symmetric for a path inversion \( \mathcal{P}(\psi(s-s_0)) = \psi(-(s-s_0)) \) with respect to an extremum of the orbital \( \psi \). Hence, \( \mathcal{P} \) is a good symmetry of the system, and is in turn used to classify the interaction.

Henceforth we set \( s_0 = 0 \). We decompose the tip–sample force into two mutually exclusive terms \( F_{TS}(s) = F_{\text{int}}(s) + F_{\text{diss}}(s) \), which are even and odd with respect to \( \mathcal{P} \), namely \( \mathcal{P}(F_{\text{int}}) = +F_{\text{int}} \) and \( \mathcal{P}(F_{\text{diss}}) = -F_{\text{diss}} \), and which correspond to reversible potential interactions and irreversible dissipative interactions, respectively ‡.

The energy that is exchanged per cycle \( \Delta E \) is given by the hysteresis integral

\[
\Delta E = \int F_{TS} \, d\psi = \int_{-L/2}^{L/2} F_{\text{diss}}(s) \frac{d\psi}{ds} \, ds. \tag{12}
\]

As \( \Delta E \) must be odd with respect to \( \mathcal{P} \), it couples only to irreversible forces with odd-\( \mathcal{P} \) symmetry. The energy loss due to dissipation must be compensated for by a corresponding driving force \( F_0 \). Note that, similarly to the tip–sample force, only the odd part of \( F_0 \) can be used to exchange energy with the system in the course of a complete cycle.

In analogy to viscous damping, it is convenient to write the dissipative force as a product of an antisymmetric ‘velocity’ and a symmetric ‘friction coefficient’:

\[
F_{\text{diss}}(s) = \omega \sqrt{(L/4)^2 - (s - L/4)^2} \frac{d\psi}{ds} \times \Gamma(z_0, a_1, s). \tag{13}
\]

Note, however, that this is only a formal analogy and no presumptions with respect to the nature of the dissipative interaction have been made. The latter could indeed originate from a viscous drag force but it could just as well arise from a stick-slip-type friction that manifests itself as an adhesion hysteresis. The advantage of the decomposition is that \( \psi \) and \( \Gamma \) belong to the same \( \mathcal{P} \) symmetry class, and hence \( \Gamma \) can be written as an unequivocal function of \( \psi \) for a given set of experimental parameters \( z_0 \) and \( a_1 \). Henceforth, we implicitly assume that the experiment is performed for constant values of \( z_0 \) and \( a_1 \), and hence omit these parameters as arguments in the friction function. The ‘velocity’ can be defined at will as long as it is an odd function in \( s \). The

‡ Note that the forces can be associated with a generalized ‘potential’ \( F_{TS}(s) = -d/ds(\Phi_{\text{int}}(s) + \Phi_{\text{diss}}(s)) \), where the conservative ‘potential’ \( \Phi_{\text{int}}(s) = d\psi/dsU_{\text{int}}(s) \) and the dissipative ‘potential’ are odd and even functions with respect to \( \mathcal{P} \)
part, equation (3), which contains only orbital and driving force, we can separate the variational integral into the well-known reversible counterpart.\(^\text{[11]}\) The polynomials
\[
\psi = -\omega a_1 \sin(\omega t)
\]
for a harmonic orbital.

Returning to dynamic force sensing, we solve the equation of motion of the dissipative system using variational methods, as for the reversible case. However, the Fourier expansion of the tip motion now also includes odd functions with respect to time reversal
\[
\psi(t) = \sum_{n=0}^{\infty} a_n \cos(n\omega t) + \sum_{n=1}^{\infty} b_n \sin(n\omega t).
\]
We have a freedom of choice for the reference frame of the time axis whose origin is set such that the first coefficient of the sine expansion, \(b_1 = 0\), vanishes.

We invoke again a perturbative approach starting from the harmonic orbital \(\psi(t) = a_1 \cos(\omega t)\) to calculate the higher-order Fourier amplitudes. In addition, we assume a harmonic driving force \(F_0(t) = F_0 \sin(\omega t)\), which is 90° out of phase with respect to \(\psi\). The 90° phase shift between driving force and tip orbital is essential as it allows us to decouple conservative and dissipative interactions in a straightforward manner.

Because of the equivalence of \(\mathcal{P}\) and time-reversal symmetry in terms of the approximate orbital and driving force, we can separate the variational integral into the well-known reversible part, equation (3), which contains only \(\mathcal{P}\)-symmetric terms and a new antisymmetric, irreversible counterpart (\(\psi^- = \sum b_n \sin(n\omega t), \delta \Psi^- = \sum \delta b_n \sin(n\omega t)\)):
\[
\delta S_{\text{diss}} = \int_0^T [\omega_0^2 \psi^- (t) + \ddot{\psi}(t)^2 - \frac{\omega_0^2}{C} (v(s(t))\Gamma(\psi(t)) + F_0(t))] \delta \psi^- \, dt \equiv 0.
\]
Invoking the projection \(u = \cos(\omega t)\) and the variation condition \(\delta S_{\text{diss}}/\delta b_n = 0\), one obtains the analogue to equation (5) for dissipative forces:
\[
F_0 = \frac{-2a_1 \omega}{\pi} \int_{-1}^{1} \Gamma(z_0 + a_1 u) U_0(u) (1 - u^2)^{1/2} \, du
\]
and \((n \geq 2)\)
\[
b_n = \frac{2a_1 \omega}{\pi} \frac{\omega_0^2}{C(\omega_0^2 - n^2 \omega^2)} \int_{-1}^{1} \Gamma(z_0 + a_1 u) U_{n-1}(u) (1 - u^2)^{1/2} \, du
\]
where equation (16) follows from the condition \(b_1 = 0\), and where \(U_n(u) = \sin((n + 1) \cos^{-1}(u))/\sin(\cos^{-1}(u))\) denotes the \(n\)th-order Chebyshev polynomial of the second kind \([11]\). The polynomials \(U_n\) form a complete orthogonal set with respect to the metric \((1 - u^2)^{1/2}\), which weights the integrand in proportion to the local tip velocity. The first few polynomials are \(U_0 = 1, U_1 = 2u, U_2 = 4u^2 - 1, U_3 = 8u^3 - 4u\), and the higher-order polynomials are obtained from the recursion relation \(U_{n+1}(u) = 2u U_n(u) - U_{n-1}(u)\). Note that the driving force is simply proportional to a velocity average of the ‘friction coefficient’ seen by the tip.

Complementary to the reversible interaction forces, we can interpret the odd Fourier coefficients \(b_n\) of the tip motion in terms of weighted moments of a generalized friction coefficient \(\Gamma(x)\) (measured for fixed values of \(z_0\) and \(a_1\)). From these moments, one can readily reconstruct the ‘friction coefficient’ and thus the dissipative force. In analogy to equations (8) and (9), we write the ‘friction coefficient’ in terms of a Chebyshev series of the second kind
\[
\Gamma(z_0 + a_1 u) = \sum_{n=0}^{\infty} \gamma_n(z_0, a_1) U_n(u)
\]
with
\[
\gamma_n(z_0, a_1) = \frac{2}{\pi} \int_{-1}^{1} \Gamma(z_0 + a_1 u) Un(u)(1 - u^2)^{1/2} \, du.
\]  
\tag{19}

Comparison with equations (16) and (17) yields the following relation, which links the Chebyshev expansion coefficients to the Fourier coefficients \(b_n\) of the orbital \((n \geq 2)\):
\[
\gamma_{n-1}(z_0, a_1) = b_n(z_0, a_1) \frac{C}{\omega a_1} \left(1 - \frac{n^2 \omega^2}{\omega_0^2}\right)
\]
\[
= b_n(z_0, a_1) \frac{1}{\omega a_1} \left(C - n^2(C + C_{\text{eff}}^i)\right)
\]  
\tag{20}

whereas the \(\gamma_0\) term of the Chebyshev expansion is proportional to the driving force
\[
\gamma_0 = -\frac{F_0}{\omega a_1}.
\]  
\tag{21}

Thus one can reconstruct the ‘friction coefficient’ and the dissipative force law probed by the tip from an amplitude measurement of the driving force and of the quadrature components \(b_n\) of the higher harmonics of the tip motion.

### 3.2. Discussion

We first discuss adhesion hysteresis, a phenomenon frequently encountered in tapping force microscopy, where the tip makes physical contact with the sample in the course of each oscillation cycle. The important point here is that the force curves for approach and retraction are different owing to irreversible contact phenomena such as the breaking of bonds or the initiation of dislocations.

An example of such an adhesion hysteresis is shown in figure 4(a), which was measured quasi-statically using a tungsten trimer tip on a Au(111) sample \[14\]. Let us now assume that the trimer tip is mounted on a vibrating force sensor and that this tip performs an oscillatory motion of an amplitude of 1 nm about a fixed mean position at \(z_0 = 0\) nm. The corresponding dynamical conservative and dissipative interaction forces, \(F_{\text{int}} = 1/2(F_a + F_r)\) and \(F_{\text{diss}} = 1/2(F_a - F_r)\), are plotted in figures 4(b) and (c), respectively, where \(F_a\) denotes the interaction force during approach (marked ‘+’ in figure 4(a)) and \(F_r\) denotes the interaction force during retraction (marked ‘◦’ in figure 4(a)).

As experimentalists, we would like to recover the full force curve from dynamical data. As discussed above, we require (1) that the phase shift between the driving force and the fundamental oscillation is exactly 90°, (2) that the magnitude of the driving force \(F_0\) is measured in absolute units, (3) that the shift of the resonance frequency is measured, and (4) that the amplitudes, \(a_n\) and \(b_n\), of the cosine and sine harmonics are measured with phase-sensitive lock-in methods. As in the example in section 2.2, a fairly accurate representation of the conservative force \(F_{\text{int}}\) is obtained using only a few terms in the Chebyshev expansion, namely, \(f_0 \ldots f_4\), meaning that cosine harmonics of up to fourth order must be measured (see figure 4(b)).

The dissipative force is recovered from the ‘friction coefficient’ using the transformation \(F_{\text{diss}}(z_0 + a_1 u) = \omega a_1(1 - u^2)^{1/2} \Gamma(z_0 + a_1 u)\). The Chebyshev series expansion of the ‘friction coefficient’ is obtained from the amplitude of the driving force \(F_0\) and the amplitudes of the sine harmonics \(b_n\), namely, \(\gamma_0 = -F_0/(\omega a_1)\), \(\gamma_1 = -b_2/(\omega a_1)(3C + 4C_{\text{eff}}^i)\), \(\gamma_2 = \ldots\)
Figure 4. (a) Adhesion hysteresis measured using a tungsten trimer tip on a Au(111) sample: ‘+’ denotes the force measured during approach, ‘○’ denotes the one measured during retraction of the tip (adapted from [14]). (b) Conservative and (c) dissipative force corresponding to the force curve shown in (a). Dashed lines correspond to the Chebyshev series representation using five terms in the expansion.

\[-b_3/(\omega a_1)(8C + 9C_{\text{eff}}^i),\] etc (see equations (20) and (21)). The recovered dissipative force using the first five terms \(\gamma_0 \ldots \gamma_4\) is plotted in figure 4(c). Again, a similar convergence as for the conservative force is obtained. The key for the method to work efficiently is that the vibration amplitude \(a_1\) matches the range of the interaction.

We now turn to genuine viscous forces that are strictly proportional to the tip velocity. Here, the friction coefficient is an invariant material parameter which is a well-defined function
of the tip–sample distance alone. In principle, one could proceed along exactly the same lines as described above, that is, invoke a Chebyshev expansion of the friction function. In this case, however, the friction coefficient function can also be determined unequivocally from a measurement of the amplitude of the driving force as a function of the offset parameter \( z_0 \). The procedure is analogous to the recovery of conservative forces from measured frequency shift curves (see [6, 12]).

Our starting point is equation (16), which can also be regarded as a convolution product:

\[
F_0(z) = -\frac{2\omega}{\pi} \int_z^{z+2a_1} \Gamma(x) \left( 1 - \left( \frac{x-z}{a_1} - 1 \right)^2 \right)^{1/2} dx
\]  

(22)

where \( z \) denotes the distance of closest sample approach (see figure 1). The function \( \Gamma(x) \) is obtained by inverting the above convolution integral. Following [6], we assume that \( a_1 \) is large compared to the range of interaction. Correspondingly, the kernel in equation (22) is approximated by the leading square-root term at closest approach, and the integration is extended to infinity:

\[
F_0(z) = -\frac{2^{3/2} \omega}{a_1^{1/2} \pi} \int_z^{\infty} \Gamma(x)(x-z)^{1/2} dx
\]  

(23)

Invoking Laplace transforms one readily finds the corresponding inversion operation

\[
\Gamma(x) = -\frac{a_1^{1/2}}{2^{1/2} \omega} \int_x^{\infty} \frac{d^2}{dz^2} F_0(z) \delta(x-z) dz
\]  

(24)

Equations (22) and (24) form the basis for recovering the friction function \( \Gamma(x) \) from measured curves of the driving force amplitude as a function of tip–sample displacement, \( F_0(z) \), by means of iterative methods described in [12]. Note that, in general, the eigenfunctions of convolution operators are exponential functions. Hence, the inversion of equation (22) is trivial for exponentially decaying driving force curves. In the large amplitude limit one has

\[
\Gamma(x) = -\frac{(\pi a_1 \kappa^3)^{1/2}}{2^{1/2} \omega} \int \Gamma(z) \delta(x-z) dz
\]  

(25)

where \( \kappa \) denotes the exponential decay constant. For inverse power laws, on the other hand, the friction coefficient decays more rapidly than the corresponding driving force curve because of the derivative operation in equation (24). Explicitly, one has

\[
\Gamma(x) = -\frac{p(p+1)N(p)a_1^{1/2}}{2^{1/2} \omega x^{3/2}} \int \Gamma(z) \delta(x-z) dz
\]  

(26)

where \( p \) is the exponent of the measured driving force function \( F_0(z) \propto 1/z^p \), and \( N(p) \) is a normalization constant,

\[
N(p) = \int_0^{\infty} u^{-1/2}(1+u)^{-p-2} du.
\]

Hence, the friction function is proportional to \( \Gamma(x) \propto 1/x^{p+3/2} \).
4. Summary and conclusion

A first-order perturbative framework has been developed that connects physical observables in dynamic force microscopy, such as frequency shift, harmonic amplitudes, and driving force, to well-defined moments of conservative and dissipative interactions probed by the tip. The theory is invariant with respect to arbitrary offsets of the time coordinate, of the orbital coordinate, and of the associated length coordinate. The particular gauge used here was selected to avoid mathematical complications. In addition, one has a freedom of choice for the definition of a formal ‘friction coefficient’ and its associated velocity, which merely needs to be a $P$-asymmetric function. However, for the scattering moments to be orthogonal, which considerably simplifies the reconstruction of the dissipative interaction force from corresponding scattering amplitudes, the formal velocity must be proportional to the physical velocity of the tip. Also, a proper selection of the phase angle of the driving force is crucial for the decoupling of conservative and dissipative interactions. The decoupling allows one to determine the laws of interaction unequivocally in a straightforward manner. This added functionality opens up new perspectives for dynamic force microscopy. In particular, the tip–sample distance used for imaging can be defined with reference to the adhesion minimum which removes the ambiguities typically encountered in close sample proximity when using a simple feedback system with the adhesion force or the frequency shift as single control parameter. Furthermore, chemical sensitivity is expected to be enhanced owing to the capability of rapidly detecting changes of the interaction force characteristics in the conservative or the dissipative channel.

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