Nonlinear Atomic Force Microscopy: Squeezing and Skewness of Micro-Mechanical Oscillators interacting with a Surface

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We propose a two-frequency driving scheme in dynamic atomic force microscopy that maximizes the interaction time between tip and sample. Using a stochastic description of the cantilever dynamics, we predict large classical squeezing and a small amount of skewness of the tip’s phase-space probability distribution. Strong position squeezing will require close contact between tip and surface, while momentum squeezing would also be possible in the van der Waals region of the tip-surface force. Employing a generalized Caldeira-Leggett model, we predict that surface-dependent dissipative forces may be the dominant source of quantum effects and propose a procedure to isolate quantum effects from thermal fluctuations.

I. INTRODUCTION

Surfaces and interfaces, i.e., the outermost layer of atoms of a material, define how it interacts with its surroundings [1]. It is therefore critically important to study the physical and chemical properties of surfaces. For more than three decades, atomic force microscopy (AFM) [2, 3] has been an indispensable tool to explore the topography of surfaces and their electrical, magnetic and elastic properties [4]. It has been applied to study a plethora of phenomena at the nano-scale, including the measurement of forces acting on individual molecules [5], the study of biological samples [6], and nanoparticles [7]. Conventional AFM consists of a cantilever with a nanometer-sized tip that interacts with the sample while being dragged over its surface (contact mode [2]). The properties of the sample, and in particular the force between tip and sample, are measured by monitoring the motion of the cantilever. In order to minimize the damage to the sample, the cantilever can be made to rapidly tap over the sample surface while scanning. The distance to the surface is manipulated by applying an oscillating, single-frequency piezoelectric force. This mode is known as dynamic AFM [8].

Our work focuses on intermodulation AFM [9], where the cantilever is driven by a two-frequency force. We pursue several goals in this work, which highlight the advantages of dynamic AFM and shed light on the conditions and parameter settings required for it. First, we provide a theoretical description of the cantilever dynamics within the context of statistical mechanics. This enables us to study tip fluctuations, opening a path to monitor additional properties of the tip-surface interaction. We focus in particular on squeezing of thermal fluctuations, which could increase the resolution of AFM. Squeezing in AFM has been studied before within a quantum-mechanical model by Passian and Siopsis [10, 11]. Our second goal is to find a driving scheme that enhances the generation of squeezing. Since this essentially amounts to maximizing the effect of the tip-surface interaction, the principle of this scheme – maximizing the interaction time – can be applied to the measurement of other parameters as well. The generation of squeezing in AFM can be compared to classical nonlinear optics, where a coupling between different harmonics may trigger phase squeezing through the Kerr effect [12].

Our third goal is to explore under which circumstances quantum effects may be measurable in dynamic AFM. There is a growing interest in quantum effects with micromechanical resonators [13–15] or even larger objects [16, 17]. A mature experimental design such as AFM may help to progress this field. In normal operations, AFM works well within the classical regime. For this reason, we have used classical statistical mechanics for most of our results. However, we also describe the cantilever as an open quantum system [18] to distinguish quantum and classical dynamics. Of particular interest is the role of dissipative surface forces, which appear to be particularly suitable to generate quantum effects.

The paper is organized as follows. In Sec. II, we present the driving scheme to maximize the interaction time. Secs. III and IV are devoted to the description of the cantilever as a statistical system. In Sec. V, we discuss our predictions for the generation of squeezing and skewness. We expound on the physics behind squeezing generation in AFM and the influence of quantum fluctuations in Sec. VI. This is followed by a conclusion VII. Several appendices contain the details of our theoretical methods.

II. DRIVING SCHEME TO MAXIMIZE SURFACE INTERACTION

Cantilever and tip of AFM form a complex mechanical system that may include bending motion and torsion [11], but a simple harmonic oscillator model often suffices and will be used here. The tip is described in phase space as a point particle with position \( x(t) \) and momentum \( p(t) \) and its dynamics is governed by Newton’s second law,

\[
\dot{p} = -kx - \gamma Qp + F_{x\tau}(t) + F_{sf} + F_{dis}. \tag{1}
\]
After a transient period, the oscillator will settle to an elementary driven damped harmonic oscillator. In absence of the sample, this model corresponds to the form of a homogeneous force with one or two frequency components. Rate dissipation arises from the deformation of the sample surface due to the interaction with the tip. It can be modelled in different ways, for instance through Kelvin-Voigt viscoelastic dissipation, a hysteretic force, a convolution integral, or a retarded response to the sample.

In this paper, we follow Ref. and model the dissipate surface force by a position-dependent drag force, $F_{\text{dis}} = -\gamma(x)p$.

In dynamic AFM, the driving force typically takes the form of a homogeneous force with one or two frequency components. Here, we consider two angular frequency components $\omega_1$, $\omega_2$.

$$F_{\text{dr}}(t) = F_1 \sin(\omega_1 t + \phi_1) + F_2 \sin(\omega_2 t + \phi_2),$$

with $\phi_1, \phi_2$ controlling the relative phase of the force components. In absence of the sample, this model corresponds to an elementary driven damped harmonic oscillator. After a transient period, the oscillator will settle to a steady-state motion of the form

$$x_0(t) = \frac{F_1}{M\omega_1} \text{Im} \left( \frac{e^{i\omega_1 t + i\phi_1}}{Z(\omega_1)} \right) + \frac{F_2}{M\omega_2} \text{Im} \left( \frac{e^{i\omega_2 t + i\phi_2}}{Z(\omega_2)} \right),$$

where

$$Z(\omega) = \frac{\omega_0^2 - \omega^2}{\omega} + i\gamma_Q$$

is the mechanical impedance of the cantilever. $M$ denotes the reduced mass of cantilever and tip, and $\omega_0 = 2\pi f_0 = \sqrt{k/M}$ the resonance angular frequency.

The principle of the proposed driving scheme is explained in Fig. 1. In dynamic AFM, the driving force induces a periodic motion of the cantilever. If the force could be arbitrarily strong, the interaction time with the surface would be maximized for a square wave motion, where the tip spends half the time period close to the surface during each cycle. In a two-frequency driving scheme, one can maximize the time the tip spends near the surface if the two frequency components of the motion correspond to the first two Fourier coefficients of a square wave. Eq. (3) implies that this can be achieved if we pick the driving frequencies as $\omega_2 = 3\omega_1$, and the force amplitudes so that they produce the Fourier coefficients, $\frac{F_1}{\pi \omega_1} \text{Im} e^{i\phi_1} Z^{-1}(\omega_1) = \frac{h}{3}$ and $\frac{F_2}{\pi \omega_2} \text{Im} e^{i\phi_2} Z^{-1}(\omega_2) = -\frac{h}{3}$. For the choice $\omega_1 = \omega_0$, $Z(\omega_0)$ is imaginary and $Z(3\omega_0)$ is a negative real number for $\gamma_Q \ll \omega_0$. The phases then can be chosen as $\phi_1 = 0$ and $\phi_2 = \frac{\pi}{2}$. However, we will see below that the optimal choice of phases will depend on the surface interaction when the tip interacts with the sample.

### III. DYNAMICAL EQUATIONS FOR PROBABILITY MOMENTS

A real tip is not a point particle, and its motion is generally subject to thermal or quantum fluctuations, which require a probabilistic description. The position of the tip is then replaced by the mean position $\bar{x}(t) = \langle x \rangle$. Here, angle brackets denote averaging with respect to a probability distribution, which can be of classical or quantum nature. Variances are described through mean values of quadratic expressions, such as $\Delta x^2 = \langle (x - \bar{x})^2 \rangle$. More generally, one can characterize a probability distribution through its moments,

$$\Delta_{n,m} = \frac{1}{2} \left[ (x - \bar{x})^n (p - \bar{p})^m + (p - \bar{p})^m (x - \bar{x})^n \right].$$

In a classical description, the ordering of position and momentum terms is irrelevant, but we keep a symmetric ordering so that our formalism can be used for a quantum description as well. Second-order moments describe position variance $\Delta x^2 = \Delta_{2,0}$, momentum variance $\Delta p^2 = \Delta_{0,2}$, and cross-correlation $\Delta_{1,1}$. Higher order moments describe a non-Gaussian structure of the distribution. For instance, $\Delta_{3,0}$ describes the skewness in position, which is a measure for how much the maximum of the distribution differs from mean value $\bar{x}$ (see section IV).

The driving scheme introduced above has the potential to induce a large amount of squeezing and a small amount of skewness in the probability distribution. Squeezing refers to the reduction of the variance of one observable, say $\Delta x$, at the expense of increasing the variance of its conjugate variable $\Delta p$. It is most often considered in...
the quantum description of light, and it is sometimes assumed that the coupling of stretching \( \Delta p \) while squeezing \( \Delta x \) is a consequence of Heisenberg’s uncertainty principle. However, squeezing also occurs in classical probability distributions, where the coupling of stretching and squeezing is a consequence of the conservation of phase-space volume for conservative forces because of Liouville’s theorem [26].

For a point particle, the dynamical equations take the form \( \ddot{\mathbf{r}} = \mathbf{F}(\mathbf{x}) \), but for an object that is described through a probability distribution, we have \( \ddot{\mathbf{r}} = \langle \mathbf{F}(\mathbf{x}) \rangle \neq \mathbf{F}(\bar{\mathbf{x}}) \). This implies that the dynamics of mean position and momentum is generally coupled to higher-order moments. For instance, \( F(x) = cx^3 \) would couple mean momentum \( \bar{p} \) to skewness \( \Delta_{3,0} \).

For a force that is not given by a power law, the dynamical equation \( \ddot{\mathbf{r}} = \langle \mathbf{F}(\mathbf{x}) \rangle \), and the equivalent equations for all moments, will not generate a closed set of equations for mean values, variances, and skewness (see App. A and B). To overcome this problem, we make a localization approximation: we assume that the probability distribution of the oscillator is so narrow that the force varies little over the width of the distribution. In this case, the mean force can be expanded as a Taylor series around the mean position,

\[
\langle \mathbf{F}(\mathbf{x}) \rangle \approx \sum_{n=1}^{\text{max}} \frac{\mathbf{F}^{(n)}(\bar{\mathbf{x}})}{n!} ((\mathbf{x} - \bar{\mathbf{x}})^n) = \sum_{n=1}^{\text{max}} \frac{\mathbf{F}^{(n)}(\bar{\mathbf{x}})}{n!} \Delta_{n,0}. \tag{6}
\]

We have used the localization approximation to derive a coupled set of equations for mean position \( \bar{x} \) and momentum \( \bar{p} \), as well as all moments \( \Delta_{n,m} \) up to third order, \((n, m) \in \{(2, 0), (1, 1), (0, 2), (3, 0), (2, 1), (1, 2), (0, 3)\}\). The result is rather lengthy and given by Eqs. (B26)-(B34). As an example, the dynamical equation for mean momentum takes the form

\[
\ddot{\mathbf{p}} = \mathbf{F} - (\gamma Q + \gamma \bar{p}) \ddot{\mathbf{p}} + \frac{\Delta_{2,0}}{2} (F'' - \bar{p} \gamma''') - \gamma' \Delta_{1,1}
+ \frac{1}{6} \Delta_{3,0} \left( F^{(3)} - \bar{p} \gamma^{(3)} \right) - \frac{1}{2} \gamma'' \Delta_{2,1}. \tag{7}
\]

In this expression, functions \( F \) and \( \gamma \) depend on the mean position, e.g., \( F = F(\bar{x}) \). We have derived these equations from a classical stochastic theory (Fokker-Planck equation, see App. A) and from a Lindblad-type master equation for open quantum systems (App. B). The results differ by genuine quantum terms, which are proportional to \( h^2 \) and are highlighted in blue in Eqs. (B26)-(B34). In addition, there is a technical difference between both derivations that only concerns the position-dependent dissipative surface force. This difference is discussed in App. B.

### IV. TIP FLUCTUATIONS: SQUEEZING AND SKEWNESS

Suppose we know the moments \( \Delta_{n,m} \), either by measuring it or by solving the dynamical equations. Can we find the probability distribution that describes the stochastic state of the oscillator? The answer is yes, and follows from the reconstruction theorem in quantum physics [27], for instance. In App. C, we derive an approximate expression for a classical phase-space probability distribution \( \rho (\mathbf{r}) \) in terms of moments, where bold symbols \( \mathbf{r} = (x, p) \) denote phase-space vectors. A multivariate skew-normal probability distribution \( \rho (\mathbf{r}) \) can be expressed as the product of a Gaussian distribution \( \rho_0 (\mathbf{r}) \) and a second function \( \Phi \) [28],

\[
\rho (\mathbf{r}) = \rho_0 (\mathbf{r}) \Phi (\mathbf{r}) \tag{8}
\]

\[
\rho_0 (\mathbf{r}) = \frac{1}{2\pi \sigma_0^2} e^{-\frac{1}{2} (\mathbf{r} - \bar{r})^T \sigma_0^{-1} (\mathbf{r} - \bar{r})} \tag{9}
\]

\[
C = \left( \begin{array}{cc} \Delta_{2,0} & \Delta_{1,1} \\ \Delta_{1,1} & \Delta_{0,2} \end{array} \right). \tag{10}
\]

The eigenvalues of correlation matrix \( C \) are denoted by \( \sigma_1^2 \) and \( \sigma_2^2 \), where \( \sigma_1 \) corresponds to the larger and \( \sigma_2 \) to the smaller phase-space variance. If the eigenvectors of \( C \) are aligned with position and momentum, position squeezing corresponds to \( \Delta x = \sigma_2 \) and \( \Delta p = \sigma_1 > \sigma_2 \) [42]. The Gaussian factor \( \rho_0 (\mathbf{r}) \) would then have an ellipsoidal form that is stretched along the \( p \)-axis and squeezed along the \( x \)-axis. In general, however, squeezing can occur along any direction in phase space.

If only information about second-order moments is available, the Gaussian part is all that can be known about \( \rho \). Knowledge about third-order moments enables us to find the following expansion of \( \Phi (\mathbf{r}) \) around the mean position \( \bar{\mathbf{r}} = (\bar{x}, \bar{p}) \),

\[
\Phi (\mathbf{r}) = 1 + \mathbf{S} \cdot \mathbf{R} + \frac{1}{6} \sum_{i,j,k} \mu_{ijk} R_i R_j R_k + \mathcal{O} (\mathbf{R}^4) \tag{11}
\]

\[
S_i = -\frac{1}{2} \sum_{j,k} \mu_{ijk} (C^{-1})_{jk} \tag{12}
\]

\[
R = C^{-1} (\mathbf{r} - \bar{\mathbf{r}}), \tag{13}
\]

where \( \mu_{111} = \Delta_{30} \), \( \mu_{222} = \Delta_{03} \), and \( \mu_{112} = \mu_{212} = \mu_{121} = \mu_{211} = \Delta_{21} \), as well as \( \mu_{221} = \mu_{122} = \mu_{112} = \Delta_{12} \). Vector \( \mathbf{S} \) corresponds to the shift of the maximum of \( \rho \) relative to the mean position \( \bar{\mathbf{r}} \), as long as \( |\mathbf{S}| \) is much smaller than the variances of \( \rho \). The third-order term (the triple sum) generates a roughly triangular distortion of the Gaussian profile.

In the following, we will solve the dynamical equations of motion and use the reconstructed classical probability distribution to visualize the effect of the surface force on variances.
frequency driving with different sets of frequencies $\omega_1$ and $\omega_2$ have considered several different cases, including single- and two-frequency driving on- and off-resonant driving, as well as two-frequency driving with different sets of frequencies $\omega_i$ and phase factors $\phi_i$ in driving force (2). Figures 2 and 3 show the reconstructed probability distribution at the time when the tip is close to the sample during cycle 102 and 198, respectively [43]. For this specific case, we have used $\omega_1 = \omega_0$, $\omega_2 = 3\omega_0$, as well as $F_1 = 1.21$ nN and $F_2 = 950$ nN. The second force component needs to be much stronger to compensate for the fact that it drives the cantilever off-resonantly. The phases of the force were chosen as $\phi_1 = \pi$ rad and $\phi_2 = 2.67$ rad. This choice of phases is rather different from the phases presented at the end of Sec. II for reasons we will discuss below.

We have chosen to display the reconstructed probability distribution after 102 cycles, because at this time the double-peak structure of the Gaussian distribution, i.e., the triple sum in Eq. (11), are strongest. Since these distortions grow with the third power of the distance vector $R_i$ from the mean position of the tip, their effect is more pronounced far away from the center of the distribution. However, even on the outermost contour in Fig. 2, the deviation from a Gaussian shape only amounts to about 2% and is barely visible. Shift vector $S$ of Eq. (12), which quantifies skewness, is negligibly small at this time, about 0.01 pm.

After 198 cycles, strong squeezing can be observed, see Fig. 3 a). For our choice of parameters, we observe almost a pure position squeezing (i.e., the ellipse is almost vertical). Skewness has grown by a factor of 10 to about 0.15 pm. However, like triangular distortions, the effect is too small to be visible in our plots.

We found that the amount of squeezing critically depends on the choice of phase factors for the driving force. For instance, Fig. 3 b) shows the same situation, but with phase factors chosen as described in Sec. II. The reason for the strong reduction in squeezing is that the surface force has a strong influence on the double-peak structure in Fig. 1, which is key to increasing the tip-surface interaction time. In the presence of the surface, the double peak tends to become asymmetric, so that the tip is not in close contact with the surface for a longer time anymore, see Fig. 4. The reason for this strong influence is that the relative sign of the two Fourier components of the approximate square wave in Fig. 1 matters a lot for its overall shape. In the figure, the two Fourier components have opposite signs and thus cancel each other out close to the central peak. However, if they have the same sign, they add up to a more pronounced single peak.

In Fig. 4, one can see that the double-peak structure varies over time. The reason is that the relative phase of the two frequency components is affected by the surface force over time. The double peak therefore varies between an approximate square wave and a single peak. By adjusting the phase factors of the two driving force components, one can mitigate this effect to some extent and thus optimize squeezing. Another possibility would be to adjust the driving frequencies slightly, to compensate for the temporal variation of the double peak.

Finally, we have studied the effect of the dissipative surface force on squeezing by comparing the result of our full simulation Fig. 3 a) to a simulation in which the dissipative surface force is reduced by a factor of $10^{-3}$. The result is shown in Fig. 3 c) and indicates that $F_{\text{dis}}$ both suppresses squeezing and has a strong influence on the axis of the ellipse. We generally found that, without $F_{\text{dis}}$, one almost always obtains momentum squeezing rather than position squeezing.
FIG. 3: a) Contour plot of the reconstructed classical probability distribution after 198 cycles, for $\phi_1 = \pi$ rad and $\phi_2 = 2.67$ rad. b) The same for $\phi_1 = 0$ rad and $\phi_2 = \frac{\pi}{2}$. c) The same as a) but with strongly reduced dissipative surface force.

FIG. 4: Evolution of the tip’s mean position over several periods $T = 1/f_0$ for a driving force with phase factors $\phi_1 = \pi$ rad and $\phi_2 = 2.67$ rad.

VI. DISCUSSION

Our results show several general trends, which are discussed in this section.

The role of the dissipative surface force is two-fold: it reduces the amount of squeezing, and it rotates the squeezing axis so that one can have (mostly) position squeezing instead of momentum squeezing. Since the fluctuation-dissipation theorem [29, 30] predicts an increase of fluctuations if a dissipative force acts on a system, it is natural that squeezing is reduced; reducing variances simply becomes more difficult. In our dynamical equations, fluctuations are described by terms proportional to $p_{th}$ in Eq. (B30). A dissipative force can change the orientation of the squeezing axis through its momentum dependence, which enables it to counteract features of the conservative surface force that will be discussed in the next paragraph.

The conservative surface force alone tends to create momentum squeezing. We conjecture that the reason for this is that, for most of the time during one oscillation cycle, the tip is moving in the long-range van der Waals tail of the surface force. As can be seen in Fig. 5, this tail has negative curvature, i.e., the force would pull particles that are closer to the sample stronger towards the surface than particles that are further away. Consequently, a phase-space probability distribution would then tend to be stretched along the spatial axis. Since $F_{sf}$ is a conservative force, Liouville’s theorem then implies squeezing in momentum direction.

Conversely, a conservative force with positive curvature would tend to squeeze the spatial direction of a probability distribution and stretch it in momentum direction. For the DMT force shown in Fig. 5, this is the case when the tip is in close contact with the surface ($x < -10$ nm). Furthermore, our numerical results suggest that this may also be the most effective way to achieve large squeezing.
FIG. 5: Conservative surface force $F_{sf}$ as a function of cantilever position $x$. The blue curve shows the modified DMT model (D2) used in this work. The orange curve displays the original DMT model (D1).

The reason is that the coupling between $\Delta_{1,1}$ and $\Delta_{0,2}$ (the first term on the right-hand side of Eq. (B30)), which is responsible for the generation of squeezing, is proportional to the gradient of the force. The gradient of the surface force is much larger in the contact region than in the van der Waals region, so that a larger amount of squeezing can be achieved.

An alternative way to achieve position squeezing is the interplay between a conservative and a non-conservative force, such as $F_{\text{dis}}$. The details of such an interplay are involved due to the overall dynamics of the tip during a cycle, which is shown in Fig. 6. For a two-frequency driving force, the tip’s mean position follows a non-circular path. In addition, the axes of the ellipse oscillate at twice the resonance frequency. The latter effect is known from optical squeezing and follows from our perturbative treatment in App. E.

Quantum effects are generally negligible in our numerical simulations. To understand how they could be increased, we used first-order perturbation theory to study the influence of a dissipative surface force $F_{\text{dis}}$ of form (B12). We concentrate on the dissipative part because it generates terms proportional to $\hbar^2$ in the dynamical equations for variances $\Delta_{2,0}, \Delta_{1,1}, \Delta_{0,2}$, whereas the conservative surface force only introduces quantum terms for third-order moments. The first-order correction to the position variance is given by

$$\Delta^{(1)}_{20} = \gamma_0 \int_0^t dt' e^{-\gamma_0 (t-t')} e^{-\frac{1}{2} \Delta_{2,0}^2} e^{-\frac{1}{2} \Delta_{1,1}^2} e^{-\frac{1}{2} \Delta_{0,2}^2} \times$$

$$\left[ \frac{(L^4 - 16 \Delta x_{th}^4)}{8 \omega_0 x_0^2} - \frac{\gamma Q}{\omega_0} \frac{L^4}{8 \Delta x_{th}^2} \sin(2 \omega_0 (t - t')) \right] \left[ \frac{L^4 (\Delta x_{th}^2 + x_0^2) - 16 \Delta x_{th}^4}{8 \Delta x_{th}^2 x_0^2} \cos(2 \omega_0 (t - t')) \right].$$

This result applies to the case of a single-frequency driving force ($F_2 = 0$ in Eq. (2)). In this case, the unperturbed mean position (3) varies like $x_0(t) = h \cos(\omega_0 t + \phi_1)$.

In Eq. (14), all quantum terms are proportional to $L^4$. Compared to terms that are proportional to thermal fluctuations $\Delta x_{th}^4$, quantum terms are suppressed by a factor $L^4/\Delta x_{th}^4 = (\hbar \omega_0^2 k_BT)^2$. For current AFM designs and at room temperature, this is only about $10^{-15}$, but for a 10-fold increased resonance frequency and cooled to liquid helium temperatures, this ratio could be increased to $10^{-8}$. While this is still small, physicists have developed powerful methods to eliminate the effect of noise from a signal. This includes spin echoes [31], Doppler-free spectroscopy [32], force-insensitivity in atom interferometry [33], and correlation measurements in AFM [34].

Here, we propose a procedure to separate quantum fluctuations from thermal fluctuations in AFM by adjusting the phase of the driving force. We start with the observation that, if a term proportional to $L^4$ appears together with $\Delta x_{th}^4$ in one algebraic expression, it is not possible to separate these two terms. This applies to the first and third term inside the square brackets in Eq. (14). Hence, the only quantum term that can potentially be separated is the term proportional to $\sin(2 \omega_0 (t - t'))$.

To isolate this term, we first note that the first term in square brackets is constant, while the other two terms are oscillating at frequency $2 \omega_0$. Hence, the first term can be eliminated through spectral analysis.

FIG. 6: Time evolution of the reconstructed probability distribution during the full 198th cycle. Each ellipse corresponds to the probability distribution at one particular time during the cycle, and is centered around the mean position of the tip at that time. The size of the ellipse corresponds to the mean variances of the distribution in phase space. For instance, the red ellipse in Fig. 3 a) corresponds to the leftmost ellipse in this figure. For better visibility, the size of each ellipse has been increased by a factor of 5.
To eliminate the second classical term, which is proportional to \( \cos(2\omega_0(t - t')) \), we note that the exponential factor in Eq. (14) suppresses the integrand unless the tip is close to the sample, i.e., when \( \cos(\omega_0t' + \phi_1) \approx -1 \), or \( \omega_0t' + \phi_1 \approx \pi(2n + 1) \), with \( n \in \mathbb{Z} \). This is a direct consequence of the 

**VII. CONCLUSION**

We have proposed a driving scheme to increase the interaction time between tip and sample in intermodulation AFM. One tip is described as a stochastic system that can exhibit fluctuations. We derived a set of coupled dynamical equations for probability moments, which have some similarity to the BBGKY hierarchy in statistical mechanics [35]. The solution to these equations enables us to reconstruct the tip’s phase-space probability distribution.

We use the driving scheme to investigate the generation of squeezing in the classical phase space of the tip. We predict that strong position squeezing is possible if the tip is in close contact with the surface, and if the phases of the two frequency components in intermodulation AFM are chosen appropriately. In the weakly interacting van der Waals regime of the tip-surface interaction, momentum squeezing is predominant. The dissipative part of the surface force has a strong influence on amount and orientation of phase-space squeezing.

To distinguish between classical and quantum effects, we derived the dynamical equations both using a classical Fokker-Planck equation and a quantum master equation. We found that AFM is generally well described by a classical model. Quantum effects tend to be much smaller than thermally induced fluctuations, but it is possible to separate both effects by adjusting the phase of the driving force in dynamic AFM.

A particularly interesting result is that dissipative surface forces can introduce quantum dynamics already at the level of variances, whereas quantum effects induced by a conservative force are tied to third-order (or higher) probability moments. The derivation of our dynamical equations in App. B indicates that the reason is a particular feature of the dissipative force. In the model we studied, it is a position-dependent drag force that depends on two non-commuting observables: position and momentum.

We conjecture that similar effects would also occur with other models for dissipative surface forces. For instance, in the hysteretic JKR model [20], the surface force changes depending on whether the tip moves towards or away from the sample; i.e., it depends on position and on the sign of the tip’s momentum. In a retarded model [23], the dissipative surface force when the tip is at position \( x(t) \) depends on an earlier position \( x(t - t_r) \), where \( t_r \) is the retardation time of the force. In Heisenberg picture, \( x(t) \) and \( x(t - t_r) \) are generally non-commuting.

By probing the probability distribution of the tip, one can examine different Fourier components of the surface force, study its fluctuations, and enhance specific effects, similar to methods used in nonlinear optics. Future work may address the question whether our driving scheme may also be helpful to measure magnetic surface forces, or if the separation scheme for quantum effects could be useful to isolate specific classical effects as well.

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Appendix A: Classical derivation of dynamical equations based on the Fokker-Planck equation

For a position-independent drag force $-\gamma Q p$, the Fokker-Planck equation [36, 37] for a classical phase-space probability distribution $f(x, p, t)$ can be written as

$$\partial_t f = -\frac{p}{M} \partial_x f - \partial_p ((F(x, t) - \gamma_Q p)f) + \frac{g}{2} \partial_p^2 f,$$

with $g = 2M\gamma_Q k_B T$. The mean value of a classical observable $A(x, p, t)$, where we admit an explicit time dependence, is given by

$$\langle A \rangle = \int dx \, dp \, f(x, p, t) A(x, p, t). \tag{A2}$$

As above, we use the notation $\bar{x}(t) \coloneqq \langle x \rangle$ and $\bar{p}(t) \coloneqq \langle p \rangle$. The time derivative of a mean value can be expressed as

$$\frac{d}{dt} \langle A \rangle = \langle \partial_t A \rangle + \frac{1}{M} \langle p \partial_x A \rangle + \langle (F - \gamma_Q p) \partial_p A \rangle + M\gamma_Q k_B T \langle \partial_p^2 A \rangle. \tag{A3}$$

To add a position-dependent drag force $-\gamma(x)p$, we start with the Langevin equation for such a force,

$$\begin{align*}
\dot{x} &= \frac{p}{M} \\
\dot{p} &= -\gamma(x)p + \xi,
\end{align*} \tag{A4, A5}$$

where $\xi$ is a noise force. We assume $\delta$-correlated noise, for which $\langle \xi(t) \rangle = 0$ and $\langle \xi(t)\xi(t') \rangle = g(t)\delta(t - t')$. The unknown function $g(t)$ can be found using the solution for the momentum,

$$\begin{align*}
p(t) &= p(0)e^{-\Gamma(t)} + \int_0^t dt' e^{-\Gamma(t)+\Gamma(t')} \xi(t') \\
\Gamma(t) &= \int_0^t dt' \gamma(x(t')). \tag{A6, A7}
\end{align*}$$

The mean kinetic energy of the particle is given by $E = \langle p^2 \rangle/(2M)$. For large times, for which $e^{-\Gamma(t)} \approx 0$, the particle should reach its equilibrium energy $E = \frac{1}{2}k_B T$. Hence,

$$\begin{align*}
Mk_B T &= \int_0^t dt' \int_0^t dt'' e^{-2\Gamma(t)+\Gamma(t')-\Gamma(t'')} \langle \xi(t')\xi(t'') \rangle \\
&= \int_0^t dt' e^{-2\Gamma(t)+2\Gamma(t')} g(t') \\
&= \int_0^t d\tau e^{-2\Gamma(t)+2\Gamma(t-\tau)} g(t - \tau). \tag{A8, A9, A10}
\end{align*}$$

If the particle does not move too far during the relaxation process, we can use the approximation $g(t - \tau) \approx g(t)$ and $\Gamma(t - \tau) \approx \Gamma(t) - \tau\gamma(x(t))$. We can also extend the integration interval to infinity. We then obtain

$$Mk_B T \approx \frac{g(t)}{2\gamma(x(t))}. \tag{A11}$$

This implies that the factor $g$ takes the same form as for the constant drag force, with $\gamma_Q$ replaced by $\gamma(x(t))$. This is the approximation we will use below, but we emphasize that this is model-dependent. For instance, if we use instead the approximation $g(t - \tau) \approx g(t) - \tau\dot{g}(t)$, we instead obtain the equation

$$Mk_B T \approx \frac{g(t)}{2\gamma(x(t))} - \frac{\dot{g}(t)}{4\gamma^2(x(t))}, \tag{A12}$$

which has the solution

$$g(t) \approx \frac{p_0^2}{2} e^{2\gamma(x(t))} \left\{ \gamma(0) - 2 \int_0^t dt'' \gamma^2(x(t'')) e^{-2\gamma(x(t''))} \right\}. \tag{A13}$$
Obviously, the precise form of \( g(t) \) is difficult to find in a classical model. However, with the simple approximation, we obtain a reasonable agreement with the quantum model. Putting everything together, we obtain the Fokker-Planck equation

\[
\frac{d}{dt} \langle A \rangle = \langle \partial_t A \rangle + \frac{1}{M} \langle p \partial_x A \rangle + \langle (F - (\gamma Q + \gamma(x))p) \partial_p A \rangle + M k_B T \langle (\gamma Q + \gamma(x)) \partial_p^2 A \rangle. \tag{A14}
\]

To derive the classical equations of motion for the correlation functions (5), we consider observables of the form

\[
A = (x - \bar{x})^n (p - \bar{p})^m,
\]

so that

\[
\langle \partial_t A \rangle = -n \dot{x} \Delta_{n-1,m} - m \dot{p} \Delta_{n,m-1}
\tag{A15}
\]

\[
\langle p \partial_x A \rangle = n (\bar{p} \Delta_{n-1,m} + \Delta_{n-1,m+1})
\tag{A16}
\]

\[
\langle \partial_p^2 A \rangle = m(m-1) \Delta_{n,m-2}.
\tag{A17}
\]

We then obtain

\[
\Delta_{n,m} = \frac{n}{M} \Delta_{n-1,m+1} - m \gamma Q \Delta_{n,m} + \frac{m}{M} \left\{ \langle F - \gamma(x) p(x - \bar{x})^n (p - \bar{p})^{m-1} \rangle - \langle F - \gamma(x)p \rangle \Delta_{n,m-1} \right\} + M k_B T m (m - 1) \left\{ \gamma Q \Delta_{n,m-2} + \langle \gamma(x)(x - \bar{x})^n (p - \bar{p})^{m-2} \rangle \right\}. \tag{A19}
\]

To obtain a closed set of coupled dynamical equations, we assume that uncertainties in position and momentum in the tip motion will remain small, and the tip remains localized. If the forces acting on the system vary little across the extension of the probability distribution, we can expand \( F(x) \) and \( \gamma(x) \) in a Taylor series around mean position and momentum,

\[
F(x) \approx \sum_{n=1}^{n_{\text{max}}} \frac{F^{(n)}(\bar{x})}{n!} (x - \bar{x})^n \tag{A20}
\]

\[
\gamma(x) \approx \sum_{n=1}^{n_{\text{max}}} \frac{\gamma^{(n)}(\bar{x})}{n!} (x - \bar{x})^n. \tag{A21}
\]

Applying this approximation to Eq. (A19) leads to

\[
\Delta_{n,m} \approx \frac{n}{M} \Delta_{n-1,m+1} - m \gamma Q \Delta_{n,m} + M k_B T m (m - 1) \left\{ \gamma Q \Delta_{n,m-2} + \sum_{l=0}^{l_{\text{max}}} \frac{\gamma^{(l)}(\bar{x})}{l!} \Delta_{n+l,m-2} \right\} + m \sum_{l=0}^{l_{\text{max}}} \frac{1}{l!} \left\{ \left( F^{(l)}(\bar{x}) - \gamma^{(l)}(\bar{x}) \bar{p} \right) \left( \Delta_{n+l,m-1} - \Delta_{l,0} \Delta_{n,m-1} \right) - \gamma^{(l)}(\bar{x})(\Delta_{n+l,m} - \Delta_{l,1} \Delta_{n,m-1}) \right\}, \tag{A22}
\]

which corresponds to the classical part (\( \hbar \to 0 \)) of the set of dynamical equations (B26)-(B34).

**Appendix B: Derivation of dynamical equations from a quantum Lindblad master equation**

To describe the cantilever as a quantum system, we model it as a 1D quantum harmonic oscillator of mass \( M \) that moves in a potential \( V(\dot{x}) \) and is subject to noise. The density matrix \( \rho \) obeys the Master equation in Lindblad form [38],

\[
\dot{\rho} = -i \frac{\hbar}{\hbar} [\hat{H}, \rho] - \sum_k \left( \hat{j}^\dagger_k \hat{j}_k \rho + \rho \hat{j}_k^\dagger \hat{j}_k - 2 \hat{j}_k \rho \hat{j}_k^\dagger \right), \tag{B1}
\]

\[
\hat{H} = \frac{\hat{p}^2}{2M} + V(\hat{x}), \tag{B2}
\]

where \( \hat{J}_k \) are jump operators. Force

\[
F(\hat{x}) = -\nabla V = -k \dot{x} + F_{\text{dr}}(t) + F_{\text{sf}}(\hat{x}). \tag{B3}
\]
contains all conservative forces that act on the cantilever. This includes the elastic force $-kx$ of the cantilever itself, the homogeneous two-frequency driving force $F_{\text{dr}}(t)$, and the conservative part $F_{\text{cl}}$ of the force exerted by the sample surface. The time evolution of the expectation value of an operator $\hat{A}(t)$, for which we admit an explicit time dependence, is given by

$$\frac{d}{dt} \langle \hat{A} \rangle = i\langle [\hat{A}, \hat{H}] \rangle - \sum_k \left\langle [\hat{A}, \hat{J}^k] \hat{J}_k + \hat{J}^k_\dagger [\hat{J}_k, \hat{A}] \right\rangle .$$

(B4)

Our goal is to derive the equations of motion for mean Position $\bar{x}(t) = \langle \hat{x} \rangle$, mean momentum $\bar{p}(t) = \langle \hat{p} \rangle$, as well as for their variances $\Delta x$ and $\Delta p$. We define a family of symmetric correlation functions $\Delta_{n,m} = \langle \hat{A}_n \hat{A}_m \rangle$, with

$$\Delta_{n,m} = \frac{1}{2} (\delta x^n \delta p^m + \delta p^n \delta x^m)$$

(B5)

$$\delta \hat{x} = \hat{x} - \bar{x}$$

(B6)

$$\delta \hat{p} = \hat{p} - \bar{p}.$$  

(B7)

There are two prominent noise sources: (i) the damping force due to the finite quality factor $Q$ of the cantilever, and (ii) the dissipative force associated with the surface interaction. Damping of the cantilever can be modeled by adding a force $F_Q = -\gamma_Q p$ with damping constant $\gamma_Q = \frac{\hbar}{Q}$ [24]. In open quantum systems, such a friction force is described through a Caldeira-Leggett model [39] in Lindblad form, see Eq. (5.117) of Ref. [40], for instance. In this model, the jump operator takes the form

$$\hat{J}_Q = \frac{\sqrt{\gamma_Q}}{2} \left( \frac{p_\hbar}{\hbar} \hat{x} + \frac{i}{p_\hbar} \hat{p} \right),$$

(B8)

and the Hamiltonian is modified by adding a term

$$\hat{H}_Q = \frac{\gamma_Q}{4} (\hat{x} \hat{p} + \hat{p} \hat{x}).$$

(B9)

Here, $p_\hbar = 2\sqrt{Mk_B T}$ corresponds to the momentum uncertainty in thermal equilibrium. Putting this together, the full action of the Caldeira-Leggett model can be written in form of a super-operator,

$$\mathcal{L}_Q \hat{A} = -\frac{i}{\hbar} [\hat{A}, \hat{H}_Q] - [\hat{A}, \hat{J}^k_\dagger] \hat{J}_k - \hat{J}^k_\dagger [\hat{J}_k, \hat{A}]$$

(B10)

$$= -\frac{\gamma_Q}{4p_\hbar^2} \hat{p} [\hat{p}, \hat{A}] - \frac{\gamma_Q p_\hbar^2}{4\hbar^2} \left[ \hat{x}, [\hat{x}, \hat{A}] \right] - \frac{i\gamma_Q}{2\hbar} \left( [\hat{A}, \hat{x}] \hat{p} + \hat{p} \hat{A}, \hat{x} \right).$$

(B11)

To describe the dissipative part of the surface-tip interaction, we adopt the model of Ref. [24], where the dissipative part takes the form of a drag force that depends on the distance from the sample, $F_{\text{dis}} = -p\gamma(x)$. For most analytical calculations, we will keep the position-dependent dissipation rate $\gamma(x)$ general. However, for numerical evaluations, we will follow Ref. [24] and use an exponential-decay model,

$$\gamma(x) = \gamma_0 \exp \left( -\frac{x + h}{x_\gamma} \right),$$

(B12)

where $\gamma_0$ is the dissipation rate at the sample surface and $x_\gamma$ the length scale on which the dissipative force drops off. In a quantum treatment, $\hat{x}$ has to be replaced by $\hat{p}/m$. One also has to write the operator product in a symmetric way to ensure that the force operator is hermitian. We therefore seek to generate a dissipative force operator of the form

$$\hat{F}_{\text{dis}} = -\frac{1}{2} (\hat{p} \gamma(\hat{x}) + \gamma(\hat{x}) \hat{p}).$$

(B13)

We remark that this reduces to the previous case for $\gamma(x) = \gamma_Q$. We found that it is possible to describe this process through a Lindblad master equation with

$$\hat{J}_{\text{dis}} = \frac{p_\hbar \sqrt{\gamma_0}}{4\hbar} \Gamma(\hat{x}) + \frac{i}{p_\hbar \sqrt{\gamma_0}} \hat{p}$$

(B14)

$$\hat{H}_{\text{dis}} = \frac{1}{4} (\hat{p} \Gamma(\hat{x}) + \Gamma(\hat{x}) \hat{p}),$$

(B15)
where $\Gamma(x)$ is the anti-derivative of $\gamma(x)$ and $\tau_0$ a time scale that will be discussed below. Note that $\tau_0$ may be a function of time. We can then again describe the full action of the dissipative force in terms of a super-operator,

$$L_{\text{dis}} \dot{A} = -\frac{i}{\hbar} [\dot{A}, \hat{H}_{\text{dis}}] - [\dot{A}, \hat{J}_{\text{dis}}^+] \hat{J}_{\text{dis}} - \hat{J}_{\text{dis}}^+ [\hat{J}_{\text{dis}}, \dot{A}]$$

(B16)

$$= -\frac{1}{\tau_0 p_{\text{th}}^2} \left[ \hat{\rho}, [\hat{p}, \dot{A}] \right] - \frac{\tau_0 p_{\text{th}}^2}{16\hbar^2} \left[ \hat{\Gamma}, [\hat{\Gamma}, \dot{A}] \right] - i \frac{\hbar^2}{2\tau_0} \left[ [\hat{A}, \hat{\Gamma}] \hat{\rho} + \hat{p} [\hat{A}, \hat{\Gamma}] \right].$$

(B17)

We now have to evaluate the equations of motion,

$$\frac{d}{dt} \langle A \rangle = \langle \partial_t \dot{A} \rangle - i \frac{\hbar}{\tau_0} \langle [\hat{A}, \hat{H}] \rangle + \left\langle L_Q \dot{A} + L_{\text{dis}} \dot{A} \right\rangle,$$

(B18)

for the correlation functions (B5), which include an explicit time dependence in their definition through $\bar{x}(t)$ and $\bar{\rho}(t)$. We start with mean position and momentum, which obey

$$\dot{\bar{x}} = \frac{\bar{p}}{M},$$

(B19)

$$\dot{\bar{p}} = \langle \hat{F}_{\text{tot}} \rangle - \gamma_Q \bar{p},$$

(B20)

where a dot denotes the time derivative $d/dt$, and $\hat{F}_{\text{tot}} = F(\dot{x}) + \hat{F}_{\text{dis}}$. For the correlation functions, we find

$$\Delta_{n,m} = \frac{n}{M} \Delta_{n-1,m+1} - i \frac{\hbar}{4M} \left[ n(n-1) \langle [\delta \dot{x}^{n-2}, \delta \rho^n] \rangle + \frac{i}{2\hbar} \langle \delta \dot{x}^n [V, \delta \rho^m] + [V, \delta \rho^m] \delta \dot{x}^n \rangle - m(\hat{F}_{\text{tot}}) \Delta_{n,m-1} \right.$$

$$- m\gamma_Q \Delta_{n,m} + \frac{\gamma_Q p_{\text{th}}^2}{4} m(m-1) \Delta_{n-2,m} - \frac{\hbar^2}{4\tau_0 p_{\text{th}}^2} \left( \gamma_Q + \frac{4}{\tau_0} \right) \left[ \langle [\hat{\Gamma}, \Delta_{n,m}] \rangle + \frac{i}{2\hbar} \langle [\hat{\Gamma}, \Delta_{n,m}] \rho + \bar{p} [\hat{\Gamma}, \Delta_{n,m}] \rangle \right].$$

(B21)

This result is exact, but its usefulness hinges on our ability to evaluate commutators of powers of $\bar{p}$ and functions of $\dot{x}$. This is accomplished in appendix F. Result (F16) states that a commutator of the form $[V(\dot{x}), \bar{p}^n]$ can be written in terms of Euler polynomials with an argument that contains $\bar{p}$ and the derivative operator $\partial/\partial \dot{x}$, which acts on $V(\dot{x})$. In particular, if the function $V(\dot{x})$ is a polynomial, the commutator corresponds to

$$\langle [\delta \dot{x}^m, \delta \rho^n] \rangle = i \sum_{l=1}^{\min(m,n)} C_{n,m,l} h^l \Delta_{m-l,n-l},$$

(B22)

where coefficients $C_{n,m,l}$ are defined through Eq. (F17). Hence, it can be expressed in terms of correlation functions (B5). However, potential $V(\dot{x})$ and factor $\Gamma(\dot{x})$ will generally not be of a polynomial form, so that the equations of motion will not form a closed set of equations. It is therefore necessary to make approximations.

With localization approximation (A20), the expectation value of all quantities appearing in the equations of motion can be expressed in terms of correlation functions (B5). For instance,

$$\langle F(\dot{x}) \rangle = \sum_{n=1}^{n_{\text{max}}} \frac{F^{(n)}(\bar{x})}{n!} \Delta_{n,0},$$

(B23)

$$\langle F_{\text{dis}}(\dot{x}) \rangle = -\frac{1}{2} \sum_{n=1}^{n_{\text{max}}} \frac{\gamma^{(n)}(\bar{x})}{n!} \left( \bar{p} (\dot{x} - \bar{x})^n + (\dot{x} - \bar{x})^n \bar{p} \right)$$

(B24)

$$= -\sum_{n=1}^{n_{\text{max}}} \frac{\gamma^{(n)}(\bar{x})}{n!} \left( \Delta_{n,1} + \bar{p} \Delta_{n,0} \right).$$

(B25)

We note that elastic force $-kx$, cantilever damping $\gamma_Q$, and the homogeneous driving force $F_{\text{dis}}(t)$ are taken into account exactly. This is because their Taylor series terminates after the first term, so that the above approximation leaves these forces unaffected.
Using similar expansions up to third order \( n_{\text{max}} = 3 \) for all terms in the equation of motions, and treating terms \( \Delta_n, m \) as of order \( \epsilon^{n+m} \) in the deviations from mean values, we arrive at the following equations,

\[
\frac{d\dot{x}}{dt} = \frac{\ddot{p}}{M} \tag{B26}
\]

\[
\frac{dp}{dt} : \text{see Eq. (7)} \tag{B27}
\]

\[
\frac{d\Delta_{0,0}}{dt} = \frac{2}{M} \Delta_{1,1} + \frac{\hbar^2}{2p_\text{th}} \left( \gamma_Q + \frac{4}{\tau_0} \right) \tag{B28}
\]

\[
\frac{d\Delta_{1,1}}{dt} = \frac{1}{M} \Delta_{0,2} - (\gamma_Q + \gamma(x)) \Delta_{1,1} + \Delta_{2,0} (F'(\bar{x}) - \ddot{\bar{p}} \gamma'(\bar{x})) - \gamma'(\bar{x}) \Delta_{2,1} + \frac{1}{2} \Delta_{3,0} (F''(\bar{x}) - \ddot{\bar{p}} \gamma''(\bar{x})) \tag{B29}
\]

\[
\frac{d\Delta_{0,2}}{dt} = 2 (F'(\bar{x}) - \ddot{\bar{p}} \gamma'(\bar{x})) \Delta_{1,1} - 2 \gamma_Q \left( \gamma_{0,2} - \frac{p_\text{th}^2}{4} \right) - 2 \gamma(\bar{x}) \Delta_{0,2} - \frac{\hbar^2}{2} \gamma''(\bar{x}) + (F''(\bar{x}) - \ddot{\bar{p}} \gamma''(\bar{x})) \Delta_{2,1} - 2 \gamma'(\bar{x}) \Delta_{1,2} + \frac{p_\text{th}^2 \tau_0}{8} \left( \gamma^2(\bar{x}) + \Delta_{2,0} (\gamma'(\bar{x})^2 + \gamma(\bar{x}) \gamma''(\bar{x})) + \left( \gamma'(\bar{x}) \gamma''(\bar{x}) + \frac{1}{3} \gamma(\bar{x}) \gamma'''(\bar{x}) \right) \Delta_{3,0} \right) \tag{B30}
\]

\[
\frac{d\Delta_{3,0}}{dt} = \frac{3}{M} \Delta_{2,1} \tag{B31}
\]

\[
\frac{d\Delta_{2,1}}{dt} = \frac{2 \Delta_{1,2}}{M} - (\gamma_Q + \gamma(x)) \Delta_{2,1} + \Delta_{3,0} (F'(\bar{x}) - \ddot{\bar{p}} \gamma'(\bar{x})) \tag{B32}
\]

\[
\frac{d\Delta_{1,2}}{dt} = \frac{\Delta_{0,3}}{M} - 2 (\gamma_Q + \gamma(x)) \Delta_{1,2} + 2 \Delta_{2,1} (F'(\bar{x}) - \ddot{\bar{p}} \gamma'(\bar{x})) + \Delta_{2,0} \left( \frac{p_\text{th}^2 \tau_0}{4} \gamma'(\bar{x}) \gamma(x) - \hbar^2 \gamma''(\bar{x}) \right) - \hbar^2 \gamma'(\bar{x}) + \Delta_{3,0} \frac{p_\text{th}^2 \tau_0}{8} \left( \gamma'(\bar{x}) \gamma''(\bar{x}) + \gamma'(\bar{x}) \right) \tag{B33}
\]

\[
\frac{d\Delta_{0,3}}{dt} = -3 \Delta_{0,3} (\gamma_Q + \gamma(x)) + 3 \Delta_{1,2} (F'(\bar{x}) - \ddot{\bar{p}} \gamma'(\bar{x})) + \frac{1}{2} \hbar^2 (F''(\bar{x}) - \ddot{\bar{p}} \gamma''(\bar{x})) - 2 \hbar^2 \Delta_{1,1} \gamma'''(\bar{x}) + \frac{3}{8} \tau_0 \bar{p}_\text{th} (2 \Delta_{1,1} \gamma(\bar{x}) \gamma'(\bar{x}) + \Delta_{2,1} (\gamma'(\bar{x})^2 + \gamma(x) \gamma''(\bar{x}))) . \tag{B34}
\]

These equations represent the main theoretical result of this work. We have verified that, except for two types of terms, they agree with the corresponding equations for a classical model based on the Fokker-Planck equation, which is derived in appendix A. The two types of terms in which the two models differ are (i) terms that depend on \( \hbar \), and (ii) terms that depend on derivatives of \( \gamma(x) \). We will now discuss these differences.

For better identification terms of type (i), we have printed all occurrences of \( \hbar \) in blue. These terms correspond to genuine quantum dynamics. The first occurrence in Eq. (B28) corresponds to the position uncertainty relation \( \Delta x \sim \hbar / \Delta p \) if the momentum uncertainty is equal to the thermal momentum \( p_\text{th} \). The only other occurrence in the equations for the second-order variances depends on the curvature \( \gamma''(\bar{x}) \) of the dissipative part of the surface force. It is interesting to observe that applying a position-dependent dissipative force may an effective way to observe differences between classical and quantum dynamics of a localized system. By comparison, quantum dynamics induced by the conservative part of the force only appear in the dynamical equation for the momentum skewness \( \Delta_{0,3} \).

We now turn to terms of type (ii) and the role of time scale \( \tau_0 \). We start by considering the stationary solution for the case that \( F = 0 = \gamma_Q \). The equations of motion then have a quasi-stationary solution of the form \( \bar{p} = 0, \ \bar{x} \) constant, as well as

\[
\Delta p = \frac{p_\text{th}}{4} \sqrt{\frac{\tau_0}{\gamma(\bar{x})}} \tag{B35}
\]

\[
\Delta_{1,1} = \frac{p_\text{th}^2 \tau_0}{16m} \tag{B36}
\]

\[
\Delta x = \sqrt{t \left( \frac{p_\text{th}^2 \tau_0}{8M^2} + \frac{2\hbar^2}{p_\text{th}^2 \tau_0} \right)} , \tag{B37}
\]

and \( \Delta_{n, m} = 0 \) for the third-order correlation functions. If we want to ensure that the momentum uncertainty \( \Delta p \) is equal to \( p_\text{th} / 2 \) in this case, we have to set \( \tau_0 = 4 / \gamma(\bar{x}) \).

A second reason why \( \tau_0 \) should be equal to \( 4 / \gamma(\bar{x}) \) is the comparison with the classical Fokker-Planck equation (A22). In the classical limit (\( \hbar = 0 \)) and for constant \( \bar{x} \), the two sets of equations only agree if \( \tau_0 \) is chosen in this way.
However, even for $\tau_0 = 4/\gamma(\bar{x})$ and in the classical limit, the Fokker-Planck equation differs from the results above if $\gamma(\bar{x}(t))$ varies with $\bar{x}(t)$, and these are the terms of type (ii). In the quantum derivation, these terms are a direct consequence of model (B14) for inhomogeneous quantum dissipative forces. This model appears to be the only one where the jump operator $\hat{J}_{\text{dis}}$ is linear in the momentum operator. Hence, as long as the dissipative force is created by systems that are Markovian (this is the underlying assumption of the Lindblad form), the derivative terms are needed for consistency. Furthermore, as discussed in appendix A, the absence of derivatives of $\gamma(x)$ in the classical equations (A22) is merely a consequence of an approximation. A refined Fokker-Planck model would likely generate similar terms, so that we believe that the presence of type (ii) terms is physically well justified.

**Appendix C: Reconstructing a classical probability distribution from correlation functions**

We consider an $N$-dimensional phase space $\mathbb{R}^N$ with $\mathbf{r}$ denoting an element of this space. In our case, $\mathbf{r} = (x,p)$ is the 2D phase space of a particle with one degree of freedom. A probability distribution $\rho(\mathbf{r})$ is a real positive function that is normalized to unity,

$$1 = \int d^N\mathbf{r} \rho(\mathbf{r}).$$

The $n$th moment around a point $\bar{\mathbf{r}} \in \mathbb{R}^N$ is defined as

$$\mu_n = \int d^N\mathbf{r} (\mathbf{r} - \bar{\mathbf{r}})^n \rho(\mathbf{r}).$$

Here and in the following, we will employ a multi-index notation: for a tuple $n = (n_1, \cdots, n_N)$ of $k$ integer numbers, we set

$$|n| = n_1 + n_2 + \cdots + n_N \quad \text{(C3)}$$

$$n! = n_1! n_2! \cdots n_N! \quad \text{(C4)}$$

$$\mathbf{r}^n = r_1^{n_1} r_2^{n_2} \cdots r_N^{n_N} \quad \text{(C5)}$$

$$D^n f(\mathbf{r}) = \frac{\partial^{|n|} f}{\partial_1^{n_1} \cdots \partial_N^{n_N}} \quad \text{(C6)}$$

In this appendix, we answer the following question: Given the moments $\mu_n$ of a probability distribution $\rho(\mathbf{r})$, can we reconstruct $\rho$?

To describe asymmetric probability distributions, Azzalini and Valle [28] introduced a multi-variable extension of the skew-normal distribution of the form

$$\rho(\mathbf{r}) = \rho_0(\mathbf{r}) \Phi(\mathbf{r}), \quad \text{(C7)}$$

where $\rho_0$ corresponds to a Gaussian distribution (9), with $C$ a positive symmetric $N \times N$ matrix.

In Ref. [28], $\Phi$ is a specific function, but we consider $\Phi$ as an unknown function that needs to be reconstructed using the moments. We denote the moments of the Gaussian probability distribution by $\mu_n^{(0)}$ and assume that $\Phi$ possesses a Taylor expansion around $\bar{\mathbf{r}}$, which in multi-index notation takes the form

$$\Phi(\mathbf{r}) = \sum_k \frac{1}{k!} D^k \Phi(\bar{\mathbf{r}}) (\mathbf{r} - \bar{\mathbf{r}})^k. \quad \text{(C8)}$$

We then can express the moments around point $\bar{\mathbf{r}}$ as

$$\mu_n = \int d^N\mathbf{r} (\mathbf{r} - \bar{\mathbf{r}})^n \rho_0(\mathbf{r}) \sum_k \frac{1}{k!} D^k \Phi(\bar{\mathbf{r}}) (\mathbf{r} - \bar{\mathbf{r}})^k$$

$$= \sum_k \frac{1}{k!} D^k \Phi(\bar{\mathbf{r}}) \mu_n^{(0)} \mu_{n+k}. \quad \text{(C9)}$$

This corresponds to a set of linear equations that enable us to express, up to a given order, $D^k \Phi(\bar{\mathbf{r}})$ in terms of $\mu_n$. Of particular interest is the case when $\bar{\mathbf{r}}$ corresponds to the measured mean value. In this situation, moments $\mu_k$ directly correspond to the correlation function $\Delta_{n,m}$ that we study in the main part.
For a Gaussian distribution, all odd moments vanish, $\mu^{(0)}_{2n+1} = 0$. The even moments are given by

$$
\mu^{(0)}_{i_1,i_2,\ldots,i_{2n}} = \int d^N r \, \rho_0(r)(r - \bar{r})_{i_1} \cdots (r - \bar{r})_{i_{2n}}
$$

$$
= \frac{1}{(2\pi)^{N/2}|C|^2} \int d^N x \, x_{i_1} \cdots x_{i_{2n}} \, e^{-\frac{1}{2}x^T C^{-1} x}
$$

$$
= \frac{1}{2^N n!} \sum_{p} \sum_{i_{p(1)} \cdots i_{p(2n-1)} \cdots i_{p(2n)}} C_{i_{p(1)}i_{p(2)} \cdots i_{p(2n-1)}i_{p(2n)}},
$$

where the sum runs over all permutations $p$ of the indices $i_1, \ldots, i_{2n}$. Explicitly, moments 2, 4, and 6 are given by

$$
\mu^{(0)}_{i_1,i_2} = C_{i_1,i_2}
$$

$$
\mu^{(0)}_{i_1,i_2,i_3,i_4} = C_{i_1,i_2}C_{i_3,i_4} + C_{i_1,i_3}C_{i_2,i_4} + C_{i_1,i_4}C_{i_2,i_3}
$$

$$
\mu^{(0)}_{i_1,i_2,i_3,i_4,i_5,i_6} = C_{i_1,i_6}C_{i_2,i_5}C_{i_3,i_4} + C_{i_1,i_5}C_{i_2,i_6}C_{i_3,i_4} + C_{i_1,i_4}C_{i_2,i_5}C_{i_3,i_6} + C_{i_1,i_3}C_{i_2,i_5}C_{i_4,i_6} + C_{i_1,i_2}C_{i_3,i_5}C_{i_4,i_6} + C_{i_1,i_2}C_{i_3,i_6}C_{i_4,i_5}
$$

$$
+ C_{i_1,i_5}C_{i_2,i_4}C_{i_3,i_6} + C_{i_1,i_4}C_{i_2,i_3}C_{i_3,i_6} + C_{i_1,i_3}C_{i_2,i_3}C_{i_4,i_5} + C_{i_1,i_2}C_{i_3,i_3}C_{i_4,i_5} + C_{i_1,i_2}C_{i_3,i_6}C_{i_4,i_5} + C_{i_1,i_5}C_{i_2,i_4}C_{i_3,i_6} + C_{i_1,i_4}C_{i_2,i_3}C_{i_3,i_6} + C_{i_1,i_3}C_{i_2,i_3}C_{i_4,i_5} + C_{i_1,i_2}C_{i_3,i_3}C_{i_4,i_5} + C_{i_1,i_2}C_{i_3,i_6}C_{i_4,i_5}.
$$

We now use these results and Eq. (10) to find the Taylor coefficients $D^k \Phi(\bar{r})$ up to order $k = 3$. We start by writing down explicitly the first four moments. $\mu_0 = 1$ expresses normalization of probability,

$$
1 = \mu_0
$$

$$
= \Phi(\bar{r})\mu^{(0)}_0 + \frac{1}{(2)!} D^2 \Phi(\bar{r}) \mu^{(0)}_2
$$

$$
= \Phi(\bar{r}) + \sum_{i_1,i_2} \frac{1}{(i_1,i_2)!} (\partial_{i_1} \partial_{i_2} \Phi(\bar{r})) C_{i_1,i_2}.
$$

Here, notation $(i_1,i_2)!$ is a multi-index notation that is equal to $2!$ if both indices are equal, and 1 otherwise. Below, we will also use $(i_1,i_2,i_3)!$ equal to $3!$ for all three indices equal, $2!$ if only two are equal, and 1 otherwise.

Since $\bar{r}$ is equal to the mean position, $\mu_1 = 0$ by definition. Hence, the second equation becomes

$$
0 = D^1 \Phi \mu^{(0)}_2 + \frac{1}{(3)!} D^3 \Phi \mu^{(0)}_4,
$$

or with explicit indices,

$$
0 = \sum_{i_2} (\partial_{i_2} \Phi) \mu^{(0)}_{i_1,i_2} + \sum_{i_2,i_3,i_4} \frac{1}{(i_2,i_3,i_4)!} (\partial_{i_2} \partial_{i_3} \partial_{i_4} \Phi) \mu^{(0)}_{i_1,i_2,i_3,i_4}
$$

$$
= \sum_{i_2} (\partial_{i_2} \Phi) C_{i_1,i_2} + \sum_{i_2,i_3,i_4} \frac{1}{(i_2,i_3,i_4)!} (\partial_{i_2} \partial_{i_3} \partial_{i_4} \Phi) (C_{i_1,i_4}C_{i_2,i_3} + C_{i_1,i_3}C_{i_2,i_4} + C_{i_1,i_2}C_{i_3,i_4}).
$$

Since the multi-factorial and the derivatives of $\Phi$ are totally symmetric under exchange of indices $i_2,i_3,i_4$, we can reduce the second factor to a sum over a single term,

$$
0 = \sum_{i_2} (\partial_{i_2} \Phi) C_{i_1,i_2} + 3 \sum_{i_2,i_3,i_4} \frac{1}{(i_2,i_3,i_4)!} (\partial_{i_2} \partial_{i_3} \partial_{i_4} \Phi) C_{i_1,i_4}C_{i_2,i_3}.
$$

Multiplying this with $(C^{-1})_{i_1,i}$ and summing over $i_1$ yields

$$
\partial_{i} \Phi(\bar{r}) = -3 \sum_{i_2,i_3} \frac{1}{(i_2,i_3)!} (\partial_{i_2} \partial_{i_3} \bar{r}) C_{i_2,i_3}.
$$

The second-order moment equals the correlation matrix, $\mu_{i_1,i_2} = \langle (r - \bar{r})_{i_1} (r - \bar{r})_{i_2} \rangle$. The equation for this moment reads

$$
\mu_2 = \Phi(\bar{r})\mu^{(0)}_2 + \frac{1}{(2)!} D^2 \Phi(\bar{r}) \mu^{(0)}_4.
$$
Together with Eq. (C18), this provides us with a linear set of equations for the \( N^2 + 1 \) coefficients \( \Phi(\hat{r}) \) and \( D^2 \Phi(\hat{r}) \). This is easy to solve if we pick matrix \( C \) in the Gaussian distribution (9) such that \( \mu_2^{(0)} = \mu_2 \). This is accomplished for the choice \( C_{i_1 i_2} = \mu_{i_1 i_2} \). Eqs. (C18) and (C25) are then easily solved by \( \Phi(\hat{r}) = 1 \) and \( D^2 \Phi(\hat{r}) = 0 \).

The equation for the third-order moment is given by

\[
\mu_3 = D^3 \Phi \mu_4^{(0)} + \frac{1}{(3)!} D^3 \Phi \mu_6^{(0)}, \tag{C26}
\]

or with explicit indices,

\[
\mu_{i_1 i_2 i_3} = \sum_{i_4} (\partial_{i_4} \Phi) \mu_{i_1 i_2 i_3 i_4} + \sum_{i_4, i_5, i_6} \frac{1}{(145 i_5 i_6)!} (\partial_{i_4} \partial_{i_5} \partial_{i_6} \Phi) \mu_{i_1 i_2 i_3 i_4 i_5 i_6}. \tag{C27}
\]

We can exploit Eq. (C24) to turn this equation into one that only contains the third-order derivatives \( (\partial_{i_4} \partial_{i_5} \partial_{i_6} \Phi) \). The triple sum contains factors like Eq. (C16), which are very lengthy. However, the high degree of symmetry of all factors involved enables us to recude it to

\[
\mu_{i_1 i_2 i_3} = 6 \sum_{i_4, i_5, i_6} \frac{1}{(i_4 i_5 i_6)!} (\partial_{i_4} \partial_{i_5} \partial_{i_6} \Phi) C_{i_1 i_4} C_{i_2 i_5} C_{i_3 i_6}. \tag{C28}
\]

This equation is easily solved and lets us determine all Taylor coefficients up to order 3,

\[
\partial_i \partial_j \partial_k \Phi(\hat{r}) = \frac{1}{6} \sum_{i_1, i_2, i_3} \mu_{i_1 i_2 i_3} (C^{-1})_{i_1 i} (C^{-1})_{i_2 j} (C^{-1})_{i_3 k}. \tag{C29}
\]

\[
\partial_i \Phi(\hat{r}) = -\frac{1}{2} \sum_{i_1, i_2, i_3} \mu_{i_1 i_2 i_3} (C^{-1})_{i_1 i} (C^{-1})_{i_2 j} (C^{-1})_{i_3 k}. \tag{C30}
\]

The full expression for \( \Phi(\hat{r}) \) is then given by Eq. (11).

Turning to the two-dimensional case that is the subject of this study, we use correlation matrix (10), with \( \mu_2^{(0)} = \mu_2 \) and

\[
C^{-1} = \sigma_1^{-2} \sigma_2^{-2} \begin{pmatrix} \Delta_{02} & \Delta_{11} & \Delta_{01} \\ \Delta_{11} & \Delta_{20} & \Delta_{12} \end{pmatrix}. \tag{C31}
\]

Matrix \( C \) has eigenvectors and eigenvalues

\[
e_1 = \frac{1}{\sqrt{2W(W - \Delta_{02} + \Delta_{20})}} \begin{pmatrix} \Delta_{20} - \Delta_{02} + W \\ 2 \Delta_{11} \end{pmatrix}, \quad C \cdot e_1 = \sigma_1^{-2} e_1 = \frac{1}{2} (\Delta_{20} + \Delta_{02} + W) e_1 \tag{C32}
\]

\[
e_2 = \frac{1}{\sqrt{2W(W + \Delta_{02} - \Delta_{20})}} \begin{pmatrix} \Delta_{20} - \Delta_{02} - W \\ 2 \Delta_{11} \end{pmatrix}, \quad C \cdot e_2 = \sigma_2^{-2} e_2 = \frac{1}{2} (\Delta_{20} + \Delta_{02} - W) e_2 \tag{C33}
\]

\[
W = \sqrt{4\Delta_{11}^2 + (\Delta_{20} - \Delta_{02})^2}. \tag{C34}
\]

Here, \( \sigma_1, \sigma_2 \) are the variances along the directions of the eigenvectors of \( C \). The third-order moment has components \( \mu_{111} = \Delta_{30}, \mu_{222} = \Delta_{03}, \) and \( \mu_{112} = \mu_{121} = \mu_{211} = \Delta_{21} \), as well as \( \mu_{221} = \mu_{212} = \mu_{122} = \Delta_{12} \). Introducing tensor components

\[
T_1^{(1)} = \Delta_{30} + \Delta_{12} \tag{C35}
\]

\[
T_2^{(1)} = \Delta_{03} + \Delta_{21} \tag{C36}
\]

\[
T_1^{(3)} = \frac{1}{3} \Delta_{30} - \Delta_{12} \tag{C37}
\]

\[
T_2^{(3)} = -\frac{1}{3} \Delta_{03} + \Delta_{21}, \tag{C38}
\]

we can express function \( \Phi \) as

\[
\Phi(\hat{r}) = 1 + S \cdot R + \frac{|R|^2}{8} T^{(1)} \cdot R + \frac{1}{8} T^{(3)} (R_1^3 - 3R_1 R_2^2) + \frac{1}{8} T^{(3)} (3R_2 R_1^2 - R_2^3). \tag{C39}
\]

In polar coordinates, \( R_1 = R \cos \phi \) and \( R_2 = R \sin \phi \), tensors \( T^{(1)} \) and \( T^{(3)} \) describe terms that vary like \( \cos \phi \) and \( \sin 3\phi \) or \( \cos 3\phi \) and \( \sin 3\phi \), respectively. Hence, \( T^{(1)} \) describes the direction in which the extended tail of \( \rho \) points, while \( T^{(3)} \) describes deformations of a triangular shape.
Appendix D: Numerical Simulations

To solve the dynamical equations (B26)-(B34) numerically, we consider a cantilever with a resonance frequency $f_0 = \omega_0/(2\pi) = 300$ kHz, a spring constant of $M\omega_0^2 = 40.0$ N/m, and a quality factor of $Q = 400$. The cantilever starts from its equilibrium position and from thermal equilibrium at room temperature (300 K).

To model the surface force, we follow Platz et al. [24] and assume an exponentially decreasing dissipative force $F_{\text{dis}} = -\gamma(x)p$, where $\gamma(x)$ given by Eq. (B12), with $x_s = 1.5$nm and $2\pi\gamma_0/\omega_0 \approx 0.065$. For the conservative surface force, we employ a modification of the van der Waals-Derjaguin-Muller-Toropov (DMT) model. In its original form, the DMT force is given by

$$F_{\text{DMT}}(x) = \begin{cases} 
\frac{HR}{6\omega_0^2} - \frac{H}{6(\omega_0^2 + 2\pi x h)^2} & x > -h \\
\frac{HR}{6\omega_0^2} + \frac{1}{3}E^* \sqrt{R}(h - x)^{\frac{3}{2}} & x < -h 
\end{cases},$$

(D1)

where $H = 3.28 \times 10^{-17}$kg m$^2$s$^{-2}$ is the Hamaker constant, which is a measure for the van der Waals interaction energy between tip and surface. $R = 10$ nm denotes the tip radius, and $a_0 = 2.7$ nm represents the intermolecular distance. $E^*$ = 1.5 GPa is the effective stiffness of the tip-sample system. The piecewise definition of this force makes it unsuitable for our purposes, since derivatives of Eq. (D1) are not well-defined at $x = -h$. We therefore employ a modified model, which is continuously differentiable,

$$F_{\text{sl}}(x) = -\frac{1.15HR}{12} \left(1 + \tanh \left(\frac{x + h}{L_f}\right)\right) + \frac{1}{2} \left(1 - \tanh \left(\frac{x + h}{L_f}\right)\right) \left(-\frac{1.15HR}{6(L_f^2 + a_0^2)} + \frac{4}{3}E^* \sqrt{R}(L_f^2 + (h + x)^2)^{\frac{3}{2}}\right).$$

(D2)

Here, $L_f = a_0/4$ controls the smoothness of the transition between van der Waals and surface region. A plot of model force (D2) and the original DMT model (D1) is shown in Fig. 5.

We have performed a series of numerical simulations of the full dynamical equations (B26)-(B34) with the parameters for the surface-tip interaction as given above. In all simulations, we have considered several special cases:

- **Full equations**: The full set of equations (B26)-(B34) is simulated
- **Variance limit**: Only second order correlation functions $\Delta_{20}, \Delta_{11}, \Delta_{02}$ are taken into account; third-order variances ($\Delta_{n,m}$ with $n + m = 3$) are set to zero.
- **Point-particle limit**: all correlation functions $\Delta_{nm}$ are assumed to vanish.
- **Reduced dissipative force**: To study the influence of the dissipative force, we have run the simulations in a situation where the dissipative surface force is reduced by a factor of $10^{-3}$.
- **No quantum terms**: All quantum terms (the blue terms in Eqns. (B26)-(B34)) are set to zero.

In addition, we have performed numerical simulations that include fourth-order correlation functions ($\Delta_{n,m}$ with $n + m = 4$) to verify that these terms can be ignored. These results were affirmative and are not presented in this paper.

In agreement with first-order perturbation theory (see appendix E), we found two general results in our simulations. (i) If one is only interested in studying mean position and momentum of the cantilever, the point-particle limit is appropriate. Variances only have a small effect on their dynamics. (ii) For standard AFM parameters, quantum terms can safely be neglected. Since thermal variances are generally several orders of magnitude larger than quantum uncertainties, our results do not support claims in the literature that AFM is quantum-limited.

The simulation supports the findings that we found in perturbation theory: significant squeezing is only generated when the tip is in contact with the sample.

Appendix E: Perturbation theory of the driven cantilever

If the tip-surface interaction is sufficiently small, the surface forces can be treated as a perturbation. To derive a solution of Eqs. (B26) - (B34) to first order in perturbation theory, we assume that the unperturbed system is initially thermalized, i.e., mean position and momentum follow a stationary trajectory, and the variances correspond...
to a thermal equilibrium. The full unperturbed solution for driving force (2) is then given by the unperturbed mean position \( x_0(t) \) of Eq. (3), unperturbed mean momentum \( p_0(t) = M\dot{x}_0 \), as well as

\[
\Delta x^2_{\text{th}} := \Delta_{(20)}^{(\text{no srfc})} = \frac{k_B T}{\hbar_0^2} \left( 1 + \left( 1 + \frac{1}{Q^2} \right) \left( \frac{\hbar_0}{4k_B T} \right)^2 \right) \tag{E1}
\]

\[
\Delta_{02}^{(\text{no srfc})} = Mk_B T \left( 1 + \left( \frac{\hbar_0}{4k_B T} \right)^2 \right), \tag{E2}
\]

and \( \Delta_{n,m} = 0 \) else. For a typical cantilever, the ratio of ground state energy \( \hbar_0 \) and thermal energy \( k_B T \) is in the order of \( 10^{-8} \). Here and in the following, we will therefore neglect terms of order \( \hbar^2 \) and only keep lowest-order quantum corrections. We will also neglect terms of order \( Q^{-1} \) since the quality factor is typically in the order of \( 10^2 \). With this approximation, we find the usual result for the thermal uncertainty of a classical oscillator, \( \Delta x_{\text{th}} = \sqrt{k_B T / \hbar_0^2} \).

To include the effect of surface forces, we consider the following dimensionless 9-component vector of first-order corrections,

\[
\vec{V} = \left( \frac{\Delta_{11}^{(1)}}{L^2}, \frac{\Delta_{12}^{(1)}}{\hbar}, \frac{\Delta_{13}^{(1)}}{L^3}, \frac{\Delta_{21}^{(1)}}{L}, \frac{\Delta_{12}^{(1)}}{\hbar^2}, \frac{\Delta_{13}^{(1)}}{L}, \frac{x^{(1)}}{L}, \frac{p^{(1)}}{L} \right), \tag{E3}
\]

where a superscript \( (1) \) indicates a first-order perturbation term and \( L = \sqrt{\hbar/(M\omega_0)} \) is the ground state width. The perturbative dynamical equations for this vector can be written as

\[
\partial_t \vec{V} = M \cdot \vec{V} + \vec{J}, \tag{E4}
\]

with matrix

\[
M = \begin{pmatrix}
0 & 2\omega_0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
-\omega_0 & -\gamma & \omega_0 & 0 & 0 & 0 & 0 & 0 & 0 \\
-2\omega_0 & -2\gamma & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -3\omega_0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & -\omega_0 & -\gamma & 2\omega_0 & 0 & 0 & 0 \\
0 & 0 & 0 & -2\omega_0 & -2\gamma & \omega_0 & 0 & 0 & 0 \\
0 & 0 & 0 & -3\omega_0 & 0 & 0 & -3\gamma & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & -3\omega_0 & -3\gamma & 0 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & -\omega_0 & -\gamma
\end{pmatrix}, \tag{E5}
\]

and inhomogeneity components \( J_4 = J_5 = J_8 = 0 \), and

\[
J_1 = -\frac{\hbar \gamma(x)}{8M\omega_0 \Delta x^2_{\text{th}}} \tag{E6}
\]

\[
J_2 = -\frac{\Delta x^2_{\text{th}} (F''_d(x_0) - p_0 \gamma'(x_0)) - \hbar (F''_d(x_0) - p_0 \gamma'(x_0) + M\gamma \gamma(x_0))}{16M^2 \omega_0^2 \Delta x^2_{\text{th}}} \tag{E7}
\]

\[
J_3 = -\frac{2M\omega_0 \Delta x^2_{\text{th}} (\gamma'(x_0)^2 + \gamma(x_0) \gamma''(x_0))}{\hbar \gamma(x_0)} + \frac{\hbar (\gamma Q (F''_d(x_0) - p_0 \gamma'(x_0)) + M\omega_0^2 \gamma(x_0))}{8M^2 \omega_0^3 \Delta x^2_{\text{th}}} - \frac{\hbar (\gamma'(x_0)^2 - 3\gamma(x_0) \gamma''(x_0))}{8M\omega_0 \gamma(x_0)} \tag{E8}
\]

\[
J_6 = -\frac{4\Delta x^2_{\text{th}} \gamma'(x_0)}{L^3} + L \Delta x^3_{\text{th}} \gamma^{(3)}(x_0) + \frac{3}{4} L \gamma'(x_0) \tag{E9}
\]

\[
J_7 = \frac{3L \gamma \gamma'(x_0)}{4\omega_0} - \frac{L (F''_d(x_0) - p_0 \gamma''(x_0))}{2M\omega_0} \tag{E10}
\]

\[
J_9 = -\frac{L}{h} (F''_d(x_0) - p_0 \gamma(x_0) + \Delta x^2_{\text{th}} (F''_d(x_0) - p_0 \gamma''(x_0))) \tag{E11}
\]

It is worthwhile to note that matrix \( M \) is block-diagonal and only couples correlation functions \( \Delta_{nm} \) of the same order \( n + m \). Hence, squeezing and other modifications of correlation functions must be generated through the inhomogeneity \( \vec{J}(t) \).

The solution of Eq. (E4) for \( \vec{V}(0) = 0 \) is given by

\[
\vec{V}(t) = \int_0^t dt' e^{M(t-t')} \cdot \vec{J}(t'). \tag{E12}
\]
This is best evaluated by using the eigenvalues of $M$. This matrix is not hermitian, but it is not singular. We can therefore express any vector in the form

$$\vec{J}(t') = \sum_{\alpha=1}^{9} \vec{c}_\alpha \vec{J}_\alpha(t'),$$

(E13)

where $\vec{c}_\alpha$ are the eigenvectors of $M$. Specifically, the relationship between the original components $J_i$ and the expansion coefficients $J_\alpha$ is given by

$$\vec{J}_1 = \frac{J_9(2\omega_0 - i\gamma_Q)}{2 \sqrt{2\omega_0}} - \frac{iJ_8}{\sqrt{2}}$$

(E14)

$$= -\frac{L}{\sqrt{2\hbar}} \left(1 - i \frac{\gamma_Q}{2\omega_0}\right) (F_{st}(x_0) - p_0 \gamma(x_0) + \Delta x_{th}^2 (F'_{st}(x_0) - p_0 \gamma''(x_0)))$$

(E15)

$$\vec{J}_2 = \frac{J_2 \gamma_Q}{\sqrt{2\omega_0}} + \frac{J_3}{\sqrt{2}}$$

(E16)

$$= -\sqrt{2} M \omega_0 \Delta x_{th}^2 \left(\frac{\gamma'(x_0)^2 + \gamma(x_0) \gamma''(x_0)}{\hbar \gamma(x_0)}\right) - \gamma_Q \left(\frac{\Delta x_{th}^2 - \frac{L^4}{16 \Delta x_{th}^4}}{\sqrt{2} \gamma(x_0)}\right) \frac{(F'_{st}(x_0) - p_0 \gamma'(x_0))}{8 \sqrt{2} M \omega_0 \gamma(x_0)}$$

(E17)

$$\vec{J}_4 = \frac{\sqrt{5} J_3 (\omega_0 - i\gamma_Q)}{4 \omega_0} - \frac{\sqrt{5} J_2 (\gamma_Q + 2i \omega_0)}{4 \omega_0} - \frac{1}{4} \sqrt{3} J_1$$

(E18)

$$\vec{J}_5 = \frac{\sqrt{5} J_5 (2 \omega_0 - 3i \gamma_Q)}{8 \omega_0} + \frac{\sqrt{5} J_6 (\gamma_Q + i \omega_0)}{4 \omega_0} - \frac{1}{4} \sqrt{5} J_4$$

(E19)

$$\vec{J}_6 = \frac{\sqrt{5} J_7 (2 \omega_0 - 3i \gamma_Q)}{8 \omega_0} - \frac{3 J_6 (\gamma_Q + i \omega_0)}{8 \omega_0} + \frac{3 i J_5 (5 \gamma_Q + 2i \omega_0)}{4 \omega_0}$$

(E20)

and $\vec{J}_2 = \vec{J}_1^*$, $\vec{J}_5 = \vec{J}_4^*$, $\vec{J}_7 = \vec{J}_6^*$, and $\vec{J}_9 = \vec{J}_8^*$. For $\gamma_Q \ll \omega_0$, the eigenvalues of $M$ are approximately given by

$$\lambda_\alpha \in \left\{-\frac{\gamma_Q}{2} - i \omega_0, -\frac{\gamma_Q}{2} + i \omega_0, -\gamma_Q - 2i \omega_0, -\gamma_Q + 2i \omega_0, -\frac{3 \gamma_Q}{2} - i \omega_0, -\frac{3 \gamma_Q}{2} + i \omega_0, -\frac{3 \gamma_Q}{2} - 3i \omega_0, -\frac{3 \gamma_Q}{2} + 3i \omega_0\right\},$$

(E21)

which leads to

$$\vec{V}(t) = \sum_{\alpha=1}^{9} \vec{c}_\alpha \int_{0}^{t} dt' e^{\lambda_\alpha(t-t')} \vec{J}_\alpha(t').$$

(E22)

From this expression, we can draw several conclusions.

(i) First-order perturbative effects on Mean position and momentum is described through terms involving $\vec{J}_1$ and $\vec{J}_2$ in
solution (E26). These terms are not affected by quantum effects. They are affected by thermal fluctuations through terms proportional to $\Delta x_{th}$ in Eq. (E16), but a numerical estimate shows that this influence is small, roughly in the order of $10^{-4}$. Therefore, in agreement with numerical simulations, we conclude that the point-particle approximation is appropriate if only the position of the tip is measured.

(ii) Terms involving $\tilde{J}_6$ to $\tilde{J}_9$ describe the influence of the surface force and quantum effects on skewness. Numerical simulations show that the overall size of skewness remains small, so that we do not discuss the details of this case.

(iii) The third-order expansion presented above is sufficient to describe squeezing and skewness for up to 300 cycles of the cantilever. For longer times, fourth-order terms (coupling to variances $\Delta_{nm}$ with $n + m = 4$) can have a strong influence on squeezing and skewness. We have analyzed the corresponding coupling numerically, but since it is not relevant for normal AFM time scales, we do not discuss it here.

(iv) Squeezing of second-order variances is the most interesting case, since it may be observable and exhibits the largest contributions due to quantum dynamics. Squeezing is introduced through terms involving $\tilde{J}_3$ to $\tilde{J}_5$. To first order in $\gamma_Q$, the corresponding eigenvectors of $M$ are given by

$$\tilde{e}_3 = \left(\frac{1}{\sqrt{2}} - \frac{\gamma_Q}{2\sqrt{2}\omega_0}, \frac{1}{\sqrt{2}}, 0, 0, 0, 0, 0\right),$$

$$\tilde{e}_4 = \left(-\frac{1}{\sqrt{3}}, -\frac{\gamma_Q}{\sqrt{3}\omega_0}, -\frac{1}{2\sqrt{2}\omega_0}, \frac{1}{\sqrt{3}}, 0, 0, 0, 0, 0\right),$$

and $\tilde{e}_5 = \tilde{e}_4$. In this expression, the first three components correspond to position variance, $\Delta x_1$, and momentum variance, respectively. For brevity, we will only discuss the position variance, for which Eq. (E26) yields

$$\Delta_{20}^{(1)} = \int_0^t dt' e^{\gamma_Q(t' - t)}\left[\frac{\gamma_Q}{2M\omega_0^2 \Delta x_{th}^2} (F_{st}(x_0(t')) - p_0(t')\gamma'(x_0(t'))\right]$$

$$- \left(\Delta x_{th}^4 - \frac{3L_4}{16}\right) \gamma''(x_0(t')) - \left(\frac{L^4}{16} + \Delta x_{th}^4\right) \frac{\gamma'(x_0(t'))^2}{\gamma(x_0(t'))}\right]$$

$$+ \int_0^t dt' e^{\gamma_Q(t' - t)} \cos(2\omega_0(t' - t')) \left[\Delta x_{th}^2 \left(\Delta x_{th}^4 - \frac{3L_4}{16}\right) \gamma''(x_0(t')) - \frac{L^4}{8} \gamma(x_0(t'))\right]$$

$$- \frac{\gamma_Q}{2M\omega_0^2} \left(\frac{L^4}{16} + \Delta x_{th}^4\right) (F_{st}(x_0(t')) - p_0(t')\gamma'(x_0(t'))) + \Delta x_{th}^4 \left(\frac{L^4}{16} + \Delta x_{th}^4\right) \frac{\gamma'(x_0(t'))^2}{\gamma(x_0(t'))}\right]$$

$$- \int_0^t dt' e^{\gamma_Q(t' - t)} \sin(2\omega_0(t' - t')) \left[\left(\frac{L^4}{16} + \Delta x_{th}^4\right) (F_{st}(x_0(t')) - p_0(t')\gamma'(x_0(t'))) + \frac{L^4}{8} M\gamma_Q \gamma(x_0(t'))\right]$$

This expression shows that quantum effects are generally very small. They enter through the ground state width $L$, which, at room temperature, is about a factor of $10^{-4}$ smaller than the thermal variance $\Delta x_{th}$ of the tip position.

To gain a better understanding of quantum effects, we have analyzed this expression for the special case of a single-frequency driving force ($F_2 = 0$ in Eq. (2)) oscillating at resonance frequency, $\omega_1 = \omega_0$. Furthermore, we concentrate on the effect of a dissipative surface force of the form (B12). The integral then reduces to Eq. (14). The implications of this result are discussed in the main text.

Appendix F: Commutators of functions of position and momentum

We consider functions of operator $\hat{x}$ and want to evaluate commutators of the form

$$X_n = [V(\hat{x}), \hat{p}^n].$$

We note that, since the commutator between $\delta \hat{x}$ and $\delta \hat{p}$ of Eqs. (B6) and (B7) is the same as that of $\hat{x}$ and $\hat{p}$, our results are also valid for commutators of the form $[V(\delta \hat{x}), \delta \hat{p}^n]$.

**Lemma 1:**

$$X_n = i \sum_{l=1}^n c_{n,l} \hbar^l R^{(n-l)}(V^{(l)})$$

$$R^{(m)}(f(\hat{x})) = \hat{p}^m f(\hat{x}) + f(\hat{x})\hat{p}^m,$$
with coefficients \( c_{n,l} \) that need to be determined.

**Proof:** For \( n = 1 \) we have

\[
X_1 = i\hbar c_{1,1} R^{(0)}(V^{(1)}),
\]

with \( c_{1,1} = \frac{1}{2} \). Assuming relation (F2) holds for \( n - 1 \), we obtain

\[
X_n = i \left( \frac{\hbar}{2} R^{(n-1)}(V') + \frac{1}{2} \sum_{r=1}^{n-1} c_{n-1,r} \hbar^r \left( \hat{p} R^{(n-r)}(V^{(r)}) + R^{(n-r)}(V^{(r)})\hat{p} \right) \right).
\]

Now,

\[
\hat{p} R^{(m)}(f) + R^{(m)}(f)\hat{p} = 2R^{(m+1)}(f) + \sum_{k=1}^{m} c_{m,k} \hbar^{k+1} R^{(m-k)}(f^{(k+1)}),
\]

so that,

\[
X_n = i \left( \frac{\hbar}{2} R^{(n-1)}(V') + \sum_{l=1}^{n-1} c_{n-1,l} \hbar^l R^{(n-l)}(V^{(l)}) + \frac{1}{2} \sum_{r=1}^{n-2} \sum_{k=1}^{n-1-r} c_{n-1-r,k} \hbar^{r+k+1} R^{(n-1-r-k)}(V^{(r+k+1)}) \right).
\]

Introducing the new summation index \( l = r + k + 1 \), we find

\[
X_n = i \left( \frac{\hbar}{2} R^{(n-1)}(V') + \sum_{l=1}^{n-1} c_{n-1,l} \hbar^l R^{(n-l)}(V^{(l)}) + \frac{1}{2} \sum_{l=3}^{n} \hbar^l R^{(n-l)}(V^{(l)}) \sum_{r=1}^{l-2} c_{n-1-r,l-r-1} \right)
\]

\[
= i \sum_{l=1}^{n} c_{n,l} \hbar^l R^{(n-l)}(V^{(l)}),
\]

with

\[
c_{n,1} = \frac{1}{2} + c_{n-1,1}
\]

\[
c_{n,2} = c_{n-1,2}
\]

\[
c_{n,l} = c_{n-1,l} + \frac{1}{2} \sum_{r=1}^{l-2} c_{n-1,r} c_{n-1-r,l-r-1} \quad \text{for } 3 \leq l \leq n.
\]

This completes the proof of lemma 1.

Eq. (F12) provides us with a recursion relation that can be used to determine all factors \( c_{n,l} \). We have verified that, up to \( n = 15 \), these factors correspond to coefficients of Euler polynomials \( E_n(x) \). More specifically, we found that

\[
\sum_{l=1}^{n} c_{n,l} x^{n-l} = i \left( i^n E_n(-ix) - x^n \right).
\]

We can now introduce super-operators defined by

\[
\overrightarrow{\mathcal{P}}^m V(\hat{x}) = \left( \frac{\hat{p}^m}{\hbar \frac{\partial}{\partial \hat{x}}} \right)^m V(\hat{x}),
\]

\[
\overleftarrow{\mathcal{P}}^m V(\hat{x}) = \left( \frac{1}{\hbar \frac{\partial}{\partial \hat{x}}} \right)^m V(\hat{x})\hat{p}^m,
\]

to write the commutation relations in a compact form,

\[
[V(\hat{x}),\hat{p}^n] = - \left( \hbar \frac{\partial}{\partial \hat{x}} \right)^n \left( i^n E_n(-i\overrightarrow{\mathcal{P}}) - \overrightarrow{\mathcal{P}}^n i^n E_n(-i\overleftarrow{\mathcal{P}}) + i^n E_n(-i\overrightarrow{\mathcal{P}}) - \overrightarrow{\mathcal{P}}^n \right) V(\hat{x}).
\]
It may appear strange that a derivative operator appears in the denominator of super-operators $\hat{\mathcal{P}}$. However, no negative powers of derivative operators appear in result (F16). We remark that a similar result for quadratic potentials has been proven by De Angelis and Vignat [41].

An important special case is when the function is a power, $V(x) = x^m$. It is easy to see that the (mean value of the) commutator then reduces to

$$
\langle [\hat{x}^m, \hat{p}^n] \rangle = 2i \sum_{l=1}^{\min(m,n)} \left( \frac{m!}{l!} \Delta_{m-l,n-l} \right).
$$

(F17)