## Coupling of conservative and dissipative forces in frequency-modulation atomic force microscopy

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Frequency modulation atomic force microscopy (FM-AFM) utilizes the principle of self-excitation to ensure the cantilever probe vibrates at its resonant frequency, regardless of the tip-sample interaction. Practically, this is achieved by fixing the phase difference between tip deflection and driving force at precisely 90°. This, in turn, decouples the frequency shift and excitation amplitude signals, enabling quantitative interpretation in terms of conservative and dissipative tip-sample interaction forces. In this article, we theoretically investigate the effect of phase detuning in the self-excitation mechanism on the coupling between conservative and dissipative forces in FM-AFM. We find that this coupling depends only on the relative difference in the drive and resonant frequencies far from the surface, and is thus very weakly dependent on the actual phase error particularly for high quality factors. This establishes that FM-AFM is highly robust with respect to phase detuning, and enables quantitative interpretation of the measured frequency shift and excitation amplitude, even while operating away from the resonant frequency with the use of appropriate replacements in the existing formalism. We also examine the calibration of phase shifts in FM-AFM measurements and demonstrate that the commonly used approach of minimizing the excitation amplitude can lead to significant phase detuning, particularly in liquid environments.

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Self-excitation of cantilever probes is often used in dynamic atomic force microscopy (AFM) to drive the cantilever at its resonant frequency, regardless of the tip-sample interaction.<sup>1</sup> This is achieved by exciting the cantilever with a force that is derived from its amplified and phase-shifted tip-deflection signal; setting the phase shift to 90° ensures the cantilever oscillates at its resonant frequency. Typically, the gain in this feedback loop is adjusted to ensure that the cantilever-tip oscillation amplitude is maintained constant as the tip approaches the sample, from which changes in energy dissipation can be measured. Variations in the effective stiffness of the lever are then easily measured by monitoring the change in resonant frequency using frequency detection circuitry, such as a phase-lock loop. Since frequency shifts can be measured with extreme sensitivity, and the time constant for the cantilever to reach steady oscillation using the technique is independent of its quality factor, this frequency modulation AFM (FM-AFM) approach presents an attractive methodology for ultra-sensitive measurements in both ambient and vacuum conditions.<sup>1,2</sup> The relative merits of this technique in comparison to conventional constant frequency excitation and static deflection measurements are discussed in Refs. 3 and 4.

Tip-sample forces encountered in FM-AFM measurements often contain both conservative and dissipative contributions, which can affect both the frequency of oscillation and the excitation amplitude. Provided the cantilever tipdeflection lags the driving force by precisely 90°, i.e., the cantilever is driven at its resonant frequency, these two types of forces formally decouple.<sup>5</sup> This feature enables interpretation of the measured frequency shift and excitation amplitudes in terms of conservative and dissipative force contributions. Consequently, knowledge of the effect of phase shifts that deviate from 90° is of fundamental importance in interpreting FM-AFM measurements from both a qualitative and quantitative perspective, and in assessing the robustness of this technique. To our knowledge, the only study examining the effect of such phase detuning was given in Ref. 6, which examined deviations in the frequency shift and excitation amplitude for a purely conservative interaction. The influence of phase detuning on the coupling of conservative and dissipative forces is yet to be examined, and is the primary focus of this study. We also reexamine the experimental approach commonly used for achieving a 90° phase shift, which involves adjusting the phase to minimize the excitation amplitude far from the sample.<sup>6</sup> Importantly, this minimum does not formally coincide with the 90° phase point and can thus lead to significant phase detuning particularly in liquid environments.

Provided the cantilever is driven well below its higher harmonic resonant frequencies, the cantilever motion can be described by a damping harmonic oscillator with a single degree of freedom,

$$m\frac{d^2w}{dt^2} + b\frac{dw}{dt} + kw = F_{\rm int} + F_{\rm drive},$$
 (1)

where *w* is the displacement of the cantilever tip from its unperturbed position, *k* is the stiffness of the cantilever, *m* is its effective mass,  $F_{int}$  is the interaction force experienced by the tip,  $F_{drive}$  is the driving force that excites the cantilever and *b* is the damping coefficient of the cantilever in the absence of an interaction force.

Noting that the interaction force is typically weak so that the change in effective stiffness of the cantilever is small and the cantilever motion is (approximately) harmonic, the displacement of the cantilever tip and the driving force, for an arbitrary phase lag  $\varphi$ , can be expressed as

$$w = a \sin(\omega t - \varphi), \quad F_{\text{drive}} = F_0 \sin \omega t,$$
 (2)

where *a* is the amplitude of oscillation and  $\omega$  is the driving frequency. Note that the interaction force in general modifies

the frequency  $\omega$  and driving force  $F_0$ . We define  $\omega_{set}$  to be the frequency of oscillation in the absence of an interaction force, i.e., far from the sample. Thus if  $\varphi = 90^\circ$ ,  $\omega_{set}$  coincides with  $\omega_{res}$  which is the resonant frequency of the cantilever in the absence of an interaction force.

Typically in FM-AFM measurements, the phase shift is fixed constant far from the sample. As the tip approaches and interacts with the sample, the phase shift  $\varphi$  between cantilever-tip displacement and driving force therefore remains constant. Consequently, all FM-AFM measurements of the change in frequency and excitation force are taken in reference to the frequency  $\omega_{set}$  and the drive force  $\overline{F}_0$ , respectively, far from the sample.

Importantly, the relative difference with respect to the actual resonant frequency  $\omega_{res}$  is normally not accessible, since the resonant frequency is inferred by adjusting the phase shift in the absence of an interaction force. As such, any phase error in this calibration procedure will be reflected directly in the measurement of  $\omega_{res}$ . We therefore define

$$\omega = \omega_{set} + \Delta \omega, \quad F_0 = \overline{F}_0 + \Delta F_0, \tag{3}$$

where  $\Delta \omega$  and  $\Delta F_0$  are the frequency shift and change in driving force, respectively, resulting from the tip-sample interaction. Substituting Eqs. (2) and (3) into Eq. (1) and performing a Fourier analysis then gives the required results

$$\frac{\Delta\omega}{\omega_{set}} \left(\frac{\omega_{set}}{\omega_{res}}\right)^2 + \frac{1}{2} \left\{ 1 - \left(\frac{\omega_{set}}{\omega_{res}}\right)^2 \right\} \frac{\Delta F_0}{\bar{F}_0} = I_{cons},\tag{4}$$

$$\frac{\Delta F_0}{\bar{F}_0} - \frac{\Delta \omega}{\omega_{set}} = I_{diss},$$

where

$$I_{cons} = -\frac{1}{\pi ka} \int_{-1}^{1} F_{even}(z+a(1+u)) \frac{u}{\sqrt{1-u^2}} du, \quad (5a)$$

$$I_{diss} = \frac{2}{\pi b} \int_{-1}^{1} \Delta \Gamma(z + a(1+u)) \sqrt{1 - u^2} du, \qquad (5b)$$

where we have used the property that the Fourier sine series only probes the even component of the force  $F_{\text{even}}$ , which is commonly referred to as the "conservative" component,<sup>7</sup> and that the odd component of the force can be formally expressed in terms of a generalized damping coefficient<sup>7</sup>  $F_{\text{odd}}=\Gamma(z,a,\omega,w(t))\dot{w}(t)$  for a fixed minimum tip-sample separation z, where  $\Delta\Gamma$  is the change in the generalized damping coefficient resulting from the interaction, i.e.,  $\Gamma=b+\Delta\Gamma$ . Equation (4) can be solved for the measured observables  $\Delta\omega/\omega_{set}$  and  $\Delta F_0/\overline{F}_0$ ,

$$\frac{\Delta\omega}{\omega_{set}} = I_{cons} + \frac{\omega_{res}^2 - \omega_{set}^2}{\omega_{res}^2 + \omega_{set}^2} (I_{cons} - I_{diss}),$$
(6a)

$$\frac{\Delta F_0}{\overline{F}_0} = I_{cons} + I_{diss} + \frac{\omega_{res}^2 - \omega_{set}^2}{\omega_{res}^2 + \omega_{set}^2} (I_{cons} - I_{diss}), \qquad (6b)$$

which gives the explicit coupling between the conservative  $I_{cons}$  and dissipative  $I_{diss}$  interaction force contributions.

Several important features of the coupling between conservative and dissipative forces on the measured relative frequency shift  $\Delta \omega / \omega_{set}$  and relative driving force  $\Delta F_0 / \overline{F}_0$  can be deduced immediately from Eq. (6). Importantly, we note that this coupling is primarily dependent on the relative difference between the unperturbed drive frequency  $\omega_{set}$  and unperturbed resonant frequency  $\omega_{res}$ , rather than the actual phase shift  $\varphi$ . This is particularly significant for cantilevers with high quality factors Q, such as encountered in ultra high vacuum (UHV) measurements,<sup>3</sup> as we shall now discuss.

We first consider the effects on the relative frequency shift  $\Delta \omega / \omega_{set}$ . In situations with high quality factors, large deviations of the phase from 90° may result in only minute changes in the oscillation frequency  $\omega_{set}$  from the resonant frequency  $\omega_{res}$ . Equation (6a) then immediately indicates that the resulting coupling between conservative and dissipative forces will be commensurately small. As an example, consider a typical cantilever operating in UHV conditions with a resonant frequency of  $f_{res} = \omega_{res} / (2\pi) = 150$  kHz and quality factor  $Q = 10,000.^6$  If the phase shift is set to  $\varphi = 20^\circ$ , the corresponding unperturbed oscillation frequency will be 149.979 kHz, giving

$$\frac{\omega_{res}^2 - \omega_{set}^2}{\omega_{res}^2 + \omega_{set}^2} \sim 10^{-4}.$$

Assuming conservative and dissipative forces contribute equally for the purpose of discussion, i.e.,  $I_{cons}$  and  $I_{diss}$  are of equal order (but not necessarily equal), Eq. (6a) immediately indicates that the relative contribution of the dissipative force to the frequency shift  $\Delta \omega / \omega_{set}$  will be 4 orders of magnitude smaller than that of the conservative force, despite the large phase anomaly. Clearly, as the quality factor is increased further, for example by the use of quartz oscillators,<sup>3</sup> this coupling becomes weaker for a given phase shift  $\varphi$ .

Importantly, the relative driving force  $\Delta F_0/\overline{F}_0$  always involves coupling between conservative and dissipative forces when operating on resonance, as is clear from Eq. (6b). The change in this degree of coupling again only varies with the relative difference between the unperturbed drive frequency  $\omega_{set}$  and unperturbed resonant frequency  $\omega_{res}$ . Therefore, rather than the phase being the key variable controlling this coupling, it is the relative frequency  $\omega_{set}/\omega_{res}$ ; this can be very small even for large phase detuning. These findings allow for the operation of FM-AFM measurements away from the resonance peak while minimizing the effect on the measured observables  $\Delta \omega/\omega_{set}$  and  $\Delta F_0/\overline{F}_0$ .

It is also interesting to note that the quantity  $\Delta F_0/\bar{F}_0$  $-\Delta \omega/\omega_{set}$ , Eq. (4), is completely independent of the phase shift  $\varphi$  and thus always gives the relative energy dissipated in the interaction  $I_{diss}$ , as defined by Eq. (5b). Consequently,  $\Delta F_0/\bar{F}_0 - \Delta \omega/\omega_{set}$  can be used to probe the frequency dependent nature of the energy dissipated in the interaction, provided the frequency dependence of the cantilever damping coefficient *b* is known. Furthermore, if it is desired to operate an FM-AFM specifically away from  $\varphi = 90^{\circ}$ , Eq. (4) can be used to determine the resulting conservative and dissipative interaction forces. This can be achieved using established inversion algorithms such as those presented in Refs. 7–10 that enable conversion of the measured relative frequency shift and change in excitation force to conservative and dissipative forces. By using the following replacements in the current algorithms:

$$\frac{\Delta\omega}{\omega_{res}} \to \frac{\Delta\omega}{\omega_{set}} \left(\frac{\omega_{set}}{\omega_{res}}\right)^2 + \frac{1}{2} \left\{ 1 - \left(\frac{\omega_{set}}{\omega_{res}}\right)^2 \right\} \frac{\Delta F_0}{\bar{F}_0}, \quad (7a)$$

$$\frac{\Delta F_0}{\overline{F}_0} - \frac{\Delta \omega}{\omega_{res}} \to \frac{\Delta F_0}{\overline{F}_0} - \frac{\Delta \omega}{\omega_{set}},\tag{7b}$$

these methodologies<sup>7-10</sup> are rigorously applicable, and facilitate measurement of conservative and dissipative forces as a function of frequency using the FM-AFM technique. This approach enables the FM-AFM technique to be used in a quantitative capacity in variable frequency measurements using a single cantilever, which may be particularly useful for investigating systems where the interaction force is frequency dependent.<sup>11</sup>

Finally, we reexamine the procedure commonly used to determine the  $\varphi = 90^{\circ}$  point in FM-AFM measurements. This point is typically achieved by varying the phase shift until the excitation force/amplitude is minimized, while maintaining constant tip amplitude.<sup>6</sup> This methodology implicitly assumes that the quality factor greatly exceeds unity. However, it is commonly used to set the 90° phase shift, regardless of the quality factor of the cantilever. Importantly, this methodology will not necessarily give the resonance condition  $\varphi = 90^{\circ}$ , since the frequency where the peak in the amplitude resonance curve lies does not formally coincide with the resonant frequency of the cantilever. To understand this connection, we present the relation between the phase  $\varphi$ , quality factor Q, resonant frequency  $\omega_{res}$  and drive frequency  $\omega_{set}$ , in the absence of a tip-sample interaction force

$$\cot \varphi = Q \left( \frac{\omega_{res}}{\omega_{set}} - \frac{\omega_{set}}{\omega_{res}} \right). \tag{8}$$

The frequency where the peak in the amplitude resonance curve occurs is given by  $^{12}$ 

$$\omega_{peak} = \omega_{res} \sqrt{1 - \frac{1}{2Q^2}}.$$
(9)

From Eqs. (8) and (9), we then find that if the peak frequency  $\omega_{peak}$  is used to set the 90° phase point, then the true phase shift is

$$\varphi = \tan^{-1} \left( 2Q \sqrt{1 - \frac{1}{2Q^2}} \right).$$
 (10)

Therefore, the resulting phase shift  $\varphi$  will be very close to 90° provided  $Q \ge 1$ . However, in ambient and liquid environments where the quality factor can be small, the resulting phase shift obtained from this procedure can significantly



FIG. 1. Plot of phase shift  $\varphi$  (degrees) as a function of the quality factor Q obtained by setting the drive frequency  $\omega_{set} = \omega_{peak}$ .

deviate from 90°. This property is illustrated in Fig. 1, where the resulting phase shift  $\varphi$  obtained from Eq. (10) is plotted as a function of the quality factor Q. At high quality factors we recover the expected result of  $\varphi = 90^{\circ}$ . However, for quality factors approaching unity, as is typical in liquid measurements,<sup>13,14</sup> the resulting phase shift will be significantly lower than the expected 90°.

Since the coupling between conservative and dissipative forces is dependent only on the relative difference between the resonant frequency  $\omega_{res}$  and the set frequency  $\omega_{set}$  in the absence of an interaction force, the resulting coupling will only be significant provided the difference between these two frequencies is not small. For this phase calibration procedure, Eqs. (6) become

$$\frac{\Delta\omega}{\omega_{set}} = I_{cons} + \frac{I_{cons} - I_{diss}}{4Q^2 - 1},$$
(11a)

$$\frac{\Delta F_0}{\overline{F}_0} = I_{cons} + I_{diss} + \frac{I_{cons} - I_{diss}}{4Q^2 - 1}.$$
 (11b)

While negligible coupling occurs for high quality factors  $Q \ge 1$ , significant coupling can occur for low quality factors,<sup>12</sup> and alternative calibration of the 90° phase point may be required to avoid such coupling.

One approach to determine this  $90^{\circ}$  phase point is to measure the thermal noise spectrum of the cantilever. For liquid environments, this should be performed in close proximity to the surface, but sufficiently far from the surface so that no interaction forces are present.<sup>14</sup> Fitting this noise power spectrum to the response of a damped harmonic oscillator, as in Ref. 15,

$$S(\omega) = \frac{A}{(\omega^2 - \omega_{res}^2)^2 + \frac{\omega^2 \omega_{res}^2}{O^2}},$$
 (12)

where A is a constant, enables determination of the true resonant frequency  $\omega_{res}$  and quality factor Q in liquid. The phase in the self-excitation circuit can then be adjusted so as to ensure that the cantilever oscillates at its resonant frequency  $\omega_{res}$ .

In summary, we have investigated the effect of phase detuning from  $90^{\circ}$  in FM-AFM on the coupling between conservative and dissipative forces. We found that this coupling depends only on the relative difference in the drive and resonant frequencies far from the surface, with the actual phase error not directly affecting this coupling especially for high quality factors. This finding indicates that FM-AFM is highly robust with respect to phase detuning, allowing for operation away from the true resonant frequency. A simple methodology was also presented enabling the quantitative determination of both conservative and dissipative forces regardless of the phase shift in the measurements. This allows for operation of the FM-AFM technique as a function of frequency while maintaining its quantitative abilities. We also investigated the commonly used procedure for establishing the resonance condition of the cantilever and found that it can

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lead to significant errors particularly for low quality factors, as are typical in liquid systems. An alternative procedure was proposed enabling the unequivocal determination of the resonance condition and thus ensuring true decoupling of conservative and dissipative forces.

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