

Measuring average tip-sample forces in intermittent-contact (tapping) force microscopy in air

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A method to measure the average normal force on a surface produced by intermittent-contact (tapping) force microscopy is presented. This force is determined by measuring the average deflection of a calibrated piezoresistive cantilever in intermittent contact with an oscillating active cantilever. Results obtained with this method are presented for a two-state cantilever motion where the piezolever force is higher for the state with the lower amplitude of vibration. © 2000 American Institute of Physics. [S0003-6951(00)02007-6]

Intermittent-contact force microscopy (ICFM) was developed to permit imaging of soft materials in air with high lateral resolution while using minimal lateral forces.¹ A force microscopy cantilever is vibrated close to its fundamental resonant frequency f_0 (typically 20–500 kHz) and the tip of the cantilever touches the surface briefly at one extreme of its motion. Contact with the surface is sensed by a decrease in amplitude of the cantilever response due to intermittent contact with the surface. While it is clear that many soft substances such as polymers¹ and biological molecules show much less damage when imaged in air by this method instead of contact force microscopy, the tip-surface forces involved have never been directly measured. The mathematical solutions to the nonlinear differential equations needed to calculate the forces exerted on a sample from a tapping tip can be extremely complicated even for mass-on-a-spring models for the active (tapping) cantilever.^{2–4} Predictions for these forces vary widely. In practical applications, the absence of a change in the sample properties with scanning is often taken as a evidence that the forces involved are insignificant. A quantitative understanding of the forces may help to control artifacts such as the observed dependence of inferred heights on imaging parameters.^{5–7}

The method presented here is to replace the *sample* in a commercial atomic force microscope⁸ by a piezoresistive silicon *cantilever* (Piezolever).^{9,10} The *average* tip-sample force is determined by an external circuit that detects the change in resistance when the Piezolever is intermittently contacted by a mechanically driven (active) oscillating cantilever. The z motion of the sample scanner slowly moves the Piezolever toward and away from the vibrating active cantilever. The microscope electronics records the output of a Piezolever circuit along with parameters such as the z distance moved, the amplitude of the active cantilever, and in some cases, the average deflection of the active cantilever

and/or the change in phase of the active cantilever relative to the driving signal.

Since the introduction of the ICFM technique, a number of simulations have been made to understand better the *maximum* force exerted on a fixed sample by an intermittently contacting tip.^{2,3,11–15} (Other simulations have predicted the maximum pressures exerted on a sample.^{16,17} Local pressures on the nanometer scale are even more difficult to measure experimentally than forces.) Because the contact between cantilever and surface is very abrupt and is expected to last for a time that is 2%–20% of one cycle of vibration,^{3,11,14,15,18,19} a detector to determine the maximum force must have a very fast response time. The Piezolevers available to us have resonant frequencies of less than 200 kHz and thus are unable to respond on the time scale necessary to measure the instantaneous force.²⁰ However, a measurement of the force averaged over many cycles can be made. (Bartzke *et al.*²¹ used a conventional cantilever to measure the average force exerted by a “needle sensor” oscillating at MHz frequencies where the forces were much larger.) The *average* force F_a estimated by us from the published data ranges from 1 to 10 nN.^{3,11,13,15,18} Recently, an explicit calculation of the average force as a function of separation between the surface and the cantilever has been presented for several different amplitudes.¹⁹ The existence of several oscillation states has been shown to affect the forces exerted on the sample. None of the published calculations are exactly applicable to our experimental situation, where an overall periodic motion of the surface can occur as the Piezolever deflects. As the active cantilever exerts a periodic force at a frequency above the fundamental resonance of the Piezolever, the higher modes of a rectangular cantilever may play an important role in determining the response of the piezolever.²⁰

In the present method, the active cantilever makes contact with the surface of the Piezolever opposite to the Piezolever’s tip. This surface is a buried-oxide layer used as an etch stop in the fabrication process.⁹ This surface was not chemically cleaned. X-ray photoemission measurements on the surface of one of the Piezolevers indicated a carbonaceous layer thick enough to obscure any signal from the oxide layer underneath. The Piezolever force constant was determined by contacting it with an active cantilever²² which

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was itself previously calibrated in a standard fashion from cantilevers of known geometry.²³ The force constants of the active cantilever and the Piezolever were determined in this way to about 40% absolute accuracy. The voltage applied across the Piezolever by the external, homemade Wheatstone bridge was about 1 V, giving power dissipation in the Piezolever of less than 1 mW. The Piezolever bridge output was recorded by the microscope electronics after amplification by a nominal factor of 10^4 and low-pass filtering with 3 dB point of 30 Hz. The calibration of the bridge output in terms of both deflection and force was obtained by simultaneously measuring the bridge output and a signal proportional to the deflection of a calibrated active cantilever as a function of the z distance the sample stage was translated with the two cantilevers contacting each other.

The data in Fig. 1 show a two-state behavior that produces larger intermittent contact forces in the state β characterized by a lower oscillation amplitude. The amplitude switching for this two-state behavior is more distinct than that usually obtained on a more rigid surface with a sharp tip and was not always observed in our ICFM measurements on a Piezolever. The switching between the two states occurs during both separation (filled symbols connected with dashed lines) and approach (open symbols connected by solid lines) of the two cantilevers. The force data in panel (a) are the average force measured by the Piezolever circuit; the deflection of the Piezolever can be obtained by dividing the force by the Piezolever measured spring constant of $k \sim 4$ N/m. During constant contact, the active cantilever deflects by an amount that is dependent on the ratio of the spring constants of the two cantilevers.²² The amplitude data in panel (b) (half the peak-to-peak magnitude) were determined from the manufacturer's amplitude detector⁸ with a calibration determined by the assumption that ICFM measurements under the same conditions on a rigid surface (mica or silicon) should give an amplitude versus distance slope of 1.0, as observed in previous studies.⁴ The calibration results obtained in that way were consistent with measurements of the peak-to-peak voltage measured on a digital oscilloscope, corrected for the frequency and amplitude response of the microscope electronics. The phase data in panel (c) are presented to give a qualitative comparison with experiments on conventional surfaces. The sign of the change in phase has been chosen to agree with that plotted in some theoretical works: a rising graph in the initial intermittent contact regime.^{2,19} The quantitative value was determined from the phase detection circuitry using calibration information from the manufacturer.⁸ Phase data are shown only where the amplitude is significant. The distance data was determined using the built-in detector in the scanner; the $z=0$ position was chosen arbitrarily.

The rectangular silicon active cantilever used for these data had parameters $f_0 = 259.3$ kHz, quality factor $Q \sim 300$, and $k \sim 30$ N/m; it was driven close to its resonance frequency with free amplitude ~ 75 nm. Scanning electron microscopy indicated that the tip was partially fractured with an irregular end with typical radius of curvature of about 300 nm. The Piezolever had length of about 160 μm , width of about 50 μm , with measured parameters of $k \sim 4$ N/m, $f_0 = 115.4$ kHz, $Q \sim 400$.

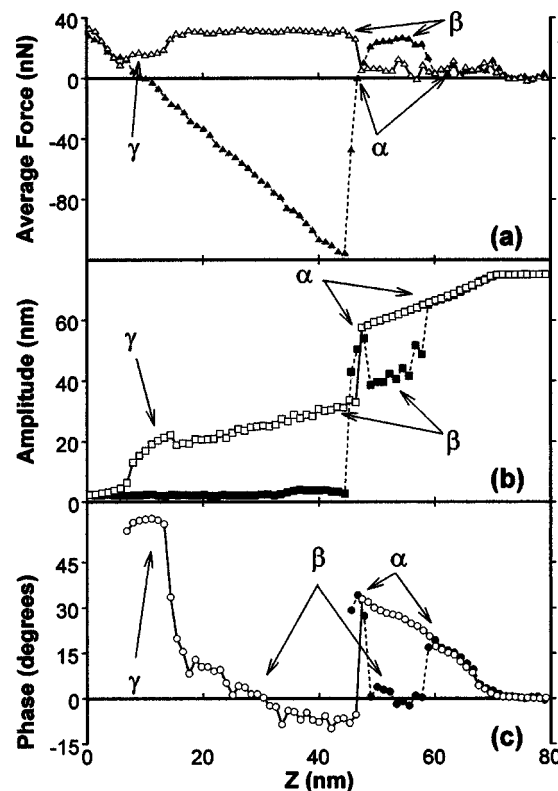


FIG. 1. Average force determined from the Piezolever deflection, the amplitude of oscillation of the active cantilever, and the phase shift of the oscillation of the active cantilever as a function of scanner position. Filled symbols connected by dashed lines are withdrawal data; open symbols connected by solid lines are approach data. The motion is seen to jump between two different states α and β ; the lower amplitude state β produces a higher force. A third state γ has an intermediate force.

The data in Fig. 1 were taken by first placing the two levers in steady contact (at a scanner position $z = -340$ nm) and then retracting the Piezolever. The dwell time at each measurement distance was 10 ms. The contact ended at $z = 45$ nm with a pull-off force of about 120 nN. (We saw this very large pull-off force for two different active cantilevers used on this Piezolever. Presumably the large force is due to the large radius of contact at the end of the tips, created by our procedure of contacting the tip to stiff surfaces for the purposes of calibrating the deflection and amplitude of the active cantilever.) The start of the intermittent-contact region is characterized by a jump in the amplitude up to $A \sim 50$ nm (state α), followed by a decrease in amplitude to $A \sim 40$ nm (state β). The average force measured by the Piezolever reaches a repulsive maximum of ~ 25 nN in those regions of the intermittent contact characterized by lower amplitude (state β). The direction of phase change when switching between the two states is consistent with the interpretation that the change in phase occurs at a switch from "attractive tapping" to "repulsive tapping" as the amplitude falls near $z = 45$ nm. The switch from the lower amplitude to the higher amplitude near $z = 60$ nm is accompanied by a decrease in the average force to a value around 5 nN (state α), close to the noise in this data set. As the sample stage is retracted further, the force remains approximately constant at about 5 nN in this case until intermittent contact ceased at about $z = 70$ nm.

After the maximum scanner separation was reached (not

shown, at $z=570$ nm in this case), the two levers were moved closer to each other. Intermittent contact occurred at about the same distance ($z=70$ nm) where it ceased on retraction, with the amplitude initially showing a gradual reduction (state α). The approach amplitude and phase curves continue past where they coincide with the retract curve until there is an abrupt jump to the low amplitude curve (state β) $z=45$ nm. A jump in phase also occurs at this point and the amplitude continues to drop until a small rise near $z=15$ nm which is accompanied by an increase in phase and a decrease in force. This is suggestive of a switch to perhaps a third state γ of less repulsive tapping. The phase behavior is similar to that observed for tapping on a polyvinyl alcohol film,²⁴ however, in that study and in simulations,¹⁹ there is a small amplitude *increase* in the intermediate region rather than *decrease* seen here.

No significant change in the average deflection of the active cantilever was detected at the sudden 25 nm drop in its amplitude in switching from state α to state β near $z=45$ nm on the approach curve. The Piezolever average position moves about 5 nm away from the active cantilever at the same time. The only way that intermittent contact can be maintained is for the Piezolever to have an oscillatory motion of at least 30 nm amplitude in state β . (We could not directly confirm this because our measurement circuit was not designed to detect the oscillatory motion of the Piezolever.) This suggests that the dynamical response of the Piezolever is significant and has to be included in any simulations that will be directly applicable to this experiment.

A wide variety of behaviors of force and amplitude versus z distance were observed. Other measurements under similar conditions (not shown) involved switching between the different states at different distances. The force measured in other data depended strongly on the amplitude and frequency of the oscillation and seemed to depend on residual impurity layers on the surface of the piezolever that was being contacted. In measurements not shown here with the same active cantilever and a different Piezolever, the force in intermittent contact was close to the noise level for our setup and the pull-off forces were much smaller. This seemed to occur only after we had scanned a 2 μ m square area of that Piezolever in contact mode; after this scanning, the impurity on the surface was moved to the perimeter of the scan area, the amplitude showed the usual behavior for a stiff surface and the average force detected by the Piezolever was very small. (The intrinsic $1/f$ noise of the Piezolever limits the smallest average force that can be measured by this method.)

The data shown here should not be taken as giving the definitive force for the given parameters because a detailed chemical characterization of the tip and the part of the Piezolever surface being contacted was not possible. Our measurements reflect the wide variation of theoretical results and indicate the extreme sensitivity of the average force to the many experimental parameters. Further studies using this technique for measuring average forces are expected. The

studies would be especially useful if coupled with simulations that include the dynamic response of the Piezolever and calculate average as well as maximum forces and pressures.

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