Dynamic Surface Force Measurement. 2. Friction and the Atomic Force Microscope

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The mechanism and geometry of force measurement with the atomic force microscope are analyzed in detail. The effective spring constant to be used in force measurement is given in terms of the cantilever spring constant. Particular attention is paid to possible dynamic effects. Theoretical calculations show that inertial effects may be neglected in most regimes, the exception being when relatively large colloidal probes are used. Model calculations of the effects of friction show that it can cause hysteresis in the constant compliance region and a shift in the zero of separation. Most surprising, friction can cause a significant diminution of the measured precontact force, and, if it actually pins the surfaces, it can change the sign of the calibration factor for the cantilever deflection, which would cause a precontact attraction to appear as a repulsion. Measurements are made of the van der Waals force between a silicon tip and a glass substrate in air. The evidence for friction and other dynamic effects is discussed. Interferometry is used to characterize the performance of the piezoelectric drive motor and position detector used in the atomic force microscope. It is shown that hysteresis in the former, and backlash in the latter, preclude a quantitative measurement of friction effects. The experimental data appear to underestimate the van der Waals attraction at high driving velocities, in qualitative agreement with the model friction calculations.

1. Introduction

In the first paper in this series, hereinafter referred to as I, certain consequences of the modern dynamic methods of surface force measurement were examined. The motivation for that paper was that with the advent of computer-controlled measurement and data acquisition techniques experiments are now performed on such fast time scales that the method of interpretation developed for the older static devices was no longer appropriate. The two main results that emerged were that inertial effects can be significant and that the dynamic decoupling of the cantilever creates uncertainty about the location of the surfaces. Detailed numeric and experimental results were obtained for the MASIF (measurement and analysis of surface forces) device.2,3

This paper deals with the AFM (atomic force microscope).4 First we apply the analysis of I to several models of the AFM in order to delimit the regime in which inertia and decoupling cannot be ignored. Second we develop and analyze several models for friction and explore the possible consequences for dynamic surface force measurement in the AFM. Third we present the results of AFM measurements of the van der Waals force in air. We characterize the AFM by interferometry and identify several artifacts of the device that have possibly in the past been confused with friction. We correct our own data for these artifacts, and we discuss particular features of the force measurements in the context of the model friction calculations.

There have been a number of previous theoretical calculations of dynamic effects, mainly in the context of noncontact or tapping mode AFM imaging.5-9 These analyses deal with a vibrating single spring and its change in amplitude due to the proximity of the surfaces. In contrast we are concerned with force measurement, not imaging, and we analyze the deflection of a realistic cantilever spring on a single approach. Our calculations go beyond the earlier studies in that in addition to the van der Waals attraction we include the moment of inertia of the cantilever, the effects of fluid drainage, and the effects of friction. Friction has been studied previously with the AFM by the lateral scanning of the tip across a substrate,10,11 and the behavior of the cantilever has been analyzed.12,13 Warmack et al.12 pointed out that friction forces cause an additional angular deflection that would appear as a change in height of the substrate. We similarly find below that friction creates uncertainty in the separation to be used for surface force measurement with the AFM. Of particular relevance to the present study is the measurements of Hoh and Engel14 who pointed out that friction can cause hysteresis in the constant compliance of surface force measurements.2,3

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2. Theoretical Modeling

2.1. Inertia. To demonstrate the regime in which inertia is significant in the AFM, we initially model the device by a sphere of mass attached to a simple spring with spring constant \( k \), interacting with a moving planar substrate at \( z(t) \). Newton’s equation of motion is

\[
mx = F_s(x) + F_d(h, h) + F_c(h)
\]

where \( x \) is the deflection of the spring and \( h = x - z(t) \) is the separation of the two surfaces. The force due to the spring is given by Hooke’s law, \( F_s = -kx \). The hydrodynamic drainage force is\(^{25} \)

\[
F_d(h, h) = -6
c

where \( \eta \) is the viscosity (\( \eta = 1.8 \times 10^{-5} \) kg m\(^{-1} \) s\(^{-1} \) in air), and \( R \) is the effective radius of curvature of the interaction region (\( R = 0 (10^2) \) nm). Drainage effects are negligible in the cases treated here, but we include this force for completeness. In this paper we take the surface force to be the van der Waals attraction in Hamaker approximation,

\[
F_c(h) = \frac{-AR}{12h^2} \left( 1 - \frac{h^6}{4R^6} \right)
\]

where \( A \) is the Hamaker constant, and \( h_0 = z_0/2^{1/3} \) is the equilibrium spacing of planar surfaces under zero load. Here we fix the soft-wall repulsion parameter at \( z_0 = 0.35 \) nm and the Hamaker constant at \( A = 5 \times 10^{-20} \) J. These parameters correspond to a surface energy of \( \gamma = A/16\pi z_0^2 = 0.08 \) J m\(^{-2} \). We set the effective radius of curvature from the measured pull-off force, which according to JKR theory is given by\(^{26} \)

\[
F_c = -3\eta R (h/4).
\]

Note that the Hamaker constant and the radius always appear as the product \( AR \), so using a smaller Hamaker constant will yield a larger effective radius of curvature. Note also that the latter is not the macroscopic diameter of the AFM or colloid probe, but rather it represents the local convexity of the contact region. The formulas that we use correspond to sphere–sphere geometry. If one is dealing with a uniformly spherical colloid interacting with a perfectly flat planar substrate, then \( R \) is half the radius of the sphere.

In \( I \) we included the effects of elastic deformation on the pre- and postcontact forces.\(^{26-28} \) These effects are much smaller for the parameters used here for the AFM and are not included in the present paper.

We used the above expressions to model typical AFM measurements of the van der Waals attraction between surfaces in air. The results are shown using representative parameters in Figure 1. The driving velocities used, 0.1–10 \( \mu \)m s\(^{-1} \), span the practical range of the device. We have chosen to model an AFM with an attached colloid probe because this is the most important configuration used for the measurement of surface forces. The 10 \( \mu \)g mass represents a relatively large probe (e.g., a melted glass capillary), and smaller colloids will obviously show smaller inertial effects; the bare cantilever itself would follow the static deflection curve on the scale of the figure (see below).

It is clear from the figure that the faster the driving velocity, the more significant are inertial effects. Quantitatively, inertia is characterized by the dimensionless parameter \( \alpha \), which is given by eq 1.6

\[
\alpha = \frac{zF_s'(h)}{4fk}\Delta_x
\]

Here \( f \) is the resonant frequency and \( \Delta_x \) is the resolution with which the deflection can be measured. Inertia is measurably significant when \( \alpha > 1 \). At a separation of \( h = 5 \) nm, a colloid of effective radius \( R = 100 \) nm has van der Waals gradient \( F_c(h) = 0.067 \) N m\(^{-1} \). With a spring constant of 0.21 N m\(^{-1} \), a 10 \( \mu \)g mass gives a resonant frequency of \( f = (k/m)^{1/2}2\pi = 0.73 \) kHz. Let us take the resolution to be \( \Delta_x = 0.5 \) nm. The fastest driving velocity used in Figure 1, \( z = 10 \mu \)m s\(^{-1} \), gives \( \alpha = 2.2 \), and the slowest driving velocity, \( z = 1 \mu \)m s\(^{-1} \), gives \( \alpha = 0.2 \). Comparing with the figure confirms that \( \alpha \approx 1 \) is a good indicator of the significance of inertial effects.

An alternative viewpoint is to consider the static jump-in point, defined by \( F_s'(h) = k \) and to estimate the inertial lag in the deflection at this point by setting \( \alpha = 1 \). These give for the inertial lag

\[
\Delta_x = \frac{z}{4f}
\]

For the fastest driving velocity of Figure 1 this is 3.4 nm, and for the slowest it is 0.3 nm, which is in reasonable agreement with the computed lag between the dynamic and static deflections at the jump-in point of the figure.

The mass of the attached colloid probe in Figure 1 is significantly larger than that of a typical bare AFM cantilever, which is of the order of nanograms. A bare
AFM cantilever (no attached colloid) has typical resonant frequencies of 20–40 kHz, which means that \( \alpha \ll 1 \) for realistic speeds and resolutions. Hence inertial effects may generally be neglected in the AFM unless a relatively large colloid probe is attached.

### 2.2. Steady-State Friction

**Figure 2.** A relatively realistic model of the AFM. The lower surface is mounted on a piezo-translator and the upper surface is mounted on a cantilever, which consists of a flexible rectangular beam of length \( L_0 \), a rigid base of length \( L_1 \), and a rigid probe at right angles to the base of length \( L_2 \), with \( L_2 \approx L_1 \approx L_0 \). The undeflected cantilever makes an angle of \( \theta_0 \approx 0^\circ \) to the horizontal. The deflection of the end of the beam is \( x \), and the angular deflection is \( \theta \), negative values corresponding to down in both cases. (The deflection is greatly exaggerated in the figure; in all cases \( x \approx L_0 \) and \( \theta \approx \theta_0 \).) The horizontal and vertical location of the tip of the probe \((y_2,z_2)\) in the coordinate system shown. For the undeflected cantilever (no forces acting), \( x = \theta = 0 \) and \( y_2 = z_2 = 0 \). The surface of the planar substrate is at \( z = 0 \), measured such that the vertical separation between the tip of the probe and the substrate is \( h = z_2 - z \). A vertical surface force \( F_z \) and a horizontal friction force \( F_x \) act on the tip of the probe.

#### 2.2.1. Effective Spring Constant

First we address the question of the force measuring spring constant. We shall begin with a simple analysis of the effect of the inclination of the cantilever, initially ignoring the tip and the base. The spring constant specified by the manufacturer, \( k_0 \), gives the resonant frequency of the cantilever. This intrinsic spring constant relates the deflection and force normal to the cantilever, \( F_0 = k_0 x \). However because the cantilever beam is inclined at an angle, one has to relate it to the force measuring spring constant, which we shall denote by \( k \) and which refers to forces and deflections normal to the substrate. The force measuring spring constant by definition is

\[
k = F_x/z_2
\]

where \( F_x \) is the force normal to the substrate acting on the tip of the probe and \( z_2 \) is the deflection of the tip, again normal to the substrate. (At this stage we assume that no friction or other horizontal force is acting, \( F_y = 0 \).) By simple geometry the component of the force \( F_x \) acting on the cantilever is \( F_x = F_0 \cos \theta_0 \) and the component of the deflection \( x \) normal to the substrate is \( z_2 = x \cos \theta_0 \). Hence

\[
k = F_x/z_2 = F_0 x \cos^2 \theta_0 = k_0 \cos^2 \theta_0
\]

For \( \theta_0 = -15^\circ \), this gives \( k = 1.07k_0 \). Failing to take the angle of inclination of the cantilever into account will lead to an underestimate of the surface force of typically 5–10%.

The cantilever spring constant is commonly measured using either resonance or gravity methods. The resonance technique(29,30) will give the intrinsic spring constant \( k_0 \), which should be corrected to the force measuring spring constant \( k \) as described above. The gravity method, where the change in deflection of the cantilever is measured via the change in the (calibrated) voltage response of the photodiode when the AFM head is inverted with a known weight on the end of the cantilever, gives the force measuring spring constant \( k_0 \) directly.

#### 2.2.2. Constant Friction

We now explore the effect of friction on the force measuring spring constant and on the calibration factor. We shall take into account the finite length of the probe and of the rigid base. Friction arises in a normal force measurement because the inclination of the cantilever causes the tip of the probe to be dragged across the substrate as the substrate is moved in the normal direction. We shall explore three models for friction, namely, when it is constant, when it is proportional to the force, and when the tip of the probe is pinned. In this section we shall assume that friction is constant.

As discussed in I, a force \( F_0 \) and a torque \( \tau \) applied to the end of the cantilever give a deflection

\[
x = \frac{1}{2B} \left[ \frac{F_0 L_0^3}{3} + \tau L_0^2 \right]
\]

and a deflection angle

\[
\theta = \frac{1}{2B} \left[ F_0 L_0^2 + 2L_0 \tau \right]
\]

Here \( B \) characterizes the flexibility of the cantilever and is related to the intrinsic spring constant by \( B = k_0 L_0^3/3 \). For a typical cantilever spring constant of \( k_0 = 0.1 \text{ N m}^{-1} \), and length \( L_0 = 200 \mu \text{m} \), the flexibility is \( B = 3 \times 10^{-13} \text{ N m}^2 \).

A vertical surface force \( F_z \), and a horizontal friction force \( F_y \), exert a force on the beam

\[
F_0 = F_z \cos \theta_0 + F_y \sin \theta_0
\]

and a torque

\[
\tau = F_z (L_1 \sin \theta_0 + L_2 \sin \theta_0) + F_y (L_1 \sin \theta_0 - L_2 \cos \theta_0)
\]

The vertical position of the tip of the probe is

\[
z_2 = x \cos \theta_0 + \left( L_1 \cos \theta_0 + L_2 \sin \theta_0 \right) \theta
\]

and its horizontal position is

\[
y_2 = x \sin \theta_0 + \left( L_1 \sin \theta_0 - L_2 \cos \theta_0 \right) \theta
\]

For brevity we shall use the obvious short-hand notation, \( \tau = a F_x + b F_y \), \( z_2 = x \cos \theta_0 + \theta \), and \( y_2 = x \sin \theta_0 + b \theta \). Note that the deflections are typically sub-microscopic, and in these equations we have retained only the leading order terms in \( x \) and \( \theta \) (e.g., \( \theta \) can be neglected compared
to \( \theta_0 \). These six equations completely specify the problem. In particular, if the friction is specified either as a constant or as a linear function of \( F_z \), then all deflections can be expressed as a linear functions of \( F_z \).

We require the force measuring spring constant \( k = \frac{dF}{dz} \) and the calibration factor \( \beta = \frac{d\theta}{dz} \). First we take \( F_y = \pm \mu \), where \( \mu > 0 \) is a constant. The positive sign is taken for loading (an inward run, \( z > 0 \)), and the negative sign for unloading. The rationale for this is that during loading, \( z > 0 \), the tip is dragged such that \( y < 0 \), and hence the frictional force must act in the opposite direction. The converse applies to unloading. The reciprocal of the force measuring spring constant is

\[
k^{-1} = \frac{dz}{dF_z} = \frac{\cos \theta_0 L_0^2}{2B \left[ 3L_0^3 \cos \theta_0 + L_0^2 a^2 \right]} + \frac{a}{2B} [L_0^2 \cos \theta_0 + 2L_0 a]
\]

(13)

If \( L_1 \) and \( L_2 \) are neglected compared to \( L_0 \), this reduces to

\[
k = \frac{3B}{L_0^3 \cos^2 \theta_0} = \frac{k_0}{\cos^2 \theta_0}
\]

(14)

in agreement with the result derived more simply above. The reciprocal of the calibration factor is

\[
\beta^{-1} = \frac{d\theta}{dz} = \frac{d\theta}{dz} = \frac{k}{2B} [L_0^2 \cos \theta_0 + 2L_0 a]
\]

(15)

It is clear that neither the spring constant nor the calibration factor are affected by a constant frictional force. However, for a given angular deflection there is a constant shift in the position that is given by

\[
z'(\theta) - z'(-\theta) = \frac{\cos \theta_0 L_0^2}{2B \left[ 3L_0^3 \cos \theta_0 + aL_0^2 \right]} \left( F_z^+ - F_z^- \right) + \mu \left[ \frac{4L_0^3}{3} \sin \theta_0 + 2bL_0 a \right]
\]

(16)

with

\[
F_z^+ - F_z^- = -2\mu L_0^2 \sin \theta_0 + 2bL_0 a
\]

(17)

That is, the inward and outward constant compliance curves will lie parallel to each other, but friction will cause the inward run to be displaced to more positive positions with respect to the outward run. Such a hysteresis is the signature of friction.

Figure 3 shows the angle-displacement curve for a loading–unloading cycle in the constant compliance regime. As the above analysis showed, the inward and outward runs lie parallel to each other but are displaced by a constant amount. In this case \( \mu = 5 \text{ nN}, k_0 = 0.13 \text{ N m}^{-1} \), the constant hysteretic offset is 0.76 nm. At \( z_2 = 10 \text{ nm} \), the inward run corresponds approximately to a force of \( F_z = 3 \text{ nN} \) and the outward run to \( F_z = 5 \text{ nN} \), which is comparable to the frictional force used, \( F_y = \pm 5 \text{ nN} \).

Obviously these quantitative values will depend on the friction coefficients of the particular surfaces. A quite reasonable model of friction would be to take it to be proportional to \( \mu \), which in the constant compliance regime would mean that \( \mu \propto z \). That is, the hysteresis shift would scale with the driving velocity.

In this case there is still a well-defined constant compliance regime (i.e., a linear region), but the gradient differs on the inward and the outward run. The friction causes hysteresis, but in this case the two constant compliance lines are no longer parallel. One should use a different spring constant and a different calibration factor on the inward and the outward runs.

Figure 4 shows the effect of friction proportional to the load, and it is noticeable in this case that the inward and outward runs do not lie parallel to each other. This is a consequence of the hysteretic offset at any point being proportional to the magnitude of the force due to friction at that point. At \( z_2 = 10 \text{ nm} \), the inward run has \( F_z = 3.4 \times 10^{-8} \text{ N} \), in the constant compliance regime. The solid lines are loading (inward run, \( z > 0 \)) and the dashed line is unloading (outward run, \( z < 0 \)). The parameters used are \( L_1 = 200 \mu \text{m}, L_2 = 4 \mu \text{m}, \theta_0 = -15^\circ \), and \( k_0 = 0.13 \text{ N m}^{-1} \).
nN, and the outward run has $F_z = 0.3 \, \text{nN}$, with the magnitude of the friction force twice as large, respectively. These can be compared with $F_y = \pm 5 \, \text{nN}$ in Figure 3. At this point the horizontal displacement is 0.65 nm. Again the quantitative results will depend on the particular surfaces that are used, but the qualitative behavior remains: the signature of the macroscopic model of friction, $F_y \propto F_x$, is that the lines of constant compliance are nonparallel.

**2.2.4. Pinning of the Tip.** The final model for friction that we shall consider is the limiting case when the tip of the probe is pinned and does not slide along the substrate. This may be an appropriate model when the adhesion is large, the surfaces are rough, or the bodies are highly deformable. In this case we assume the horizontal position to be fixed at $y_2 = 0$, which gives

$$x = -\frac{b}{\sin \theta_0} \theta$$  \hspace{1cm} (20)

It follows that the vertical position is $z_2 = \beta \theta$, with

$$\beta = (1 - \cot \theta_0)b$$  \hspace{1cm} (21)

Since in general $\theta_0 < 0$ and $b < 0$, for the case of a pinned probe the calibration factor is negative. That is, an increase in position $z_2$ leads to a decrease (more negative) deflection angle. This is the opposite sign to that of the two models analyzed above. One consequence of pinning is that an attractive force prior to contact (no pinning) will appear to be repulsive due to the negative calibration factor. The converse holds for a precontact repulsion. In the case of pinning there is no hysteresis in the inward and outward runs, except for the usual jump out of contact.

**2.3. Dynamic Friction.** In this section we shall explore the effects of friction on the dynamic measurement of surface forces. Whereas the preceding section was concerned with the steady-state motion in the constant compliance regime, here we shall incorporate friction into the equations of motion and calculate the consequent dynamic trajectory, much as was done in our exploration of inertial effects. Instead of a simple spring we shall use the full cantilever model of the AFM, including the rigid base and probe. As in I, we ascribe a mass $m_L$ uniformly distributed along the base, which is of length $L_1$, and a mass $m_0$ to the point where the base joins the cantilever. This is a rather simplified model of the AFM cantilever, since unlike the MASIF modeled in I, the mass of the cantilever is here significant. However, a full finite element analysis of the dynamics of the cantilever is beyond the scope of the present paper, and the present masses may be regarded as effective parameters that enable the general behavior of the device to be characterized.

The equations of motion are very similar to those developed in I, namely

$$m_0 \ddot{q}_a = A_{aa} q_a + A_{aq} q_q + F_z \frac{\partial h}{\partial q_a} + F_y \frac{\partial y_2}{\partial q_a}$$  \hspace{1cm} (22)

$$m_0 \ddot{q}_q = A_{aq} q_a + A_{qq} q_q + F_z \frac{\partial h}{\partial q_q} + F_y \frac{\partial y_2}{\partial q_q}$$  \hspace{1cm} (23)

Here the generalized coordinates are $q_a = x$ and $q_q = L_1 \theta + 3x/2$, and the conjugate masses are $m_0 = m_0 + m_0/4$ and $m_1 = m_0/3$. The elements of the dynamical matrix $A$ can be found in eqs. 1.17 and 1.18. The normal force is $F_z(h) = F_s(h) + F_d(h,h)$, the van der Waals and drainage forces being given above. For technical reasons we treat the latter as if it were derivable from a potential of the form $U_{\alpha}(h, \dot{h})h^b$. We have similarly treated the friction force as derivable from a potential.

From the geometry of Figure 2, as manifest in eqs 7–12, $\theta = (q_1 - 3q_2/2)/L_1$, $z_2 = q_1 \cos \theta_0 + a(q_1 - 3q_2/2)/L_1$, and $h = z_2 - z(t)$. Hence

$$\left( \frac{\partial h}{\partial q_1} \right)_{q_1} = \cos \theta_0 - \frac{3a}{2L_1}$$  \hspace{1cm} (24)

and

$$\left( \frac{\partial h}{\partial q_2} \right)_{q_2} = \frac{a}{L_1}$$  \hspace{1cm} (25)

Also, $y_2 = q_1 \sin \theta_0 + b(q_1 - 3q_2/2)L_1$, and hence

$$\left( \frac{\partial y_2}{\partial q_1} \right)_{q_1} = \sin \theta_0 - \frac{3b}{2L_1}$$  \hspace{1cm} (26)

and

$$\left( \frac{\partial y_2}{\partial q_2} \right)_{q_2} = \frac{b}{L_1}$$  \hspace{1cm} (27)

We used values of $m_0 = m_1 = 10^{-10} \, \text{kg}$, throughout, which for the usual parameters ($B = 3.5 \times 10^{-12} \, \text{N m}^2$, $L_0 = 200 \, \mu\text{m}$, $L_1 = 2 \, \mu\text{m}$, $L_2 = 4 \, \mu\text{m}$) gives a resonant frequency of 4 kHz and a spring constant of 0.13 N m$^{-1}$. This is at the low end of the resonant frequencies of many AFM cantilevers, but it provided results in reasonable computer time, and there was little evident mass dependence. We have already shown that inertial effects are negligible in a typical AFM setup, and here we focus on friction rather than inertia. We checked that the dynamic results were quantitatively unchanged when the mass was increased by a factor of 100.

The model for friction that we use in this section is

$$F_y = -\xi y_2$$  \hspace{1cm} (28)

This model, which may be traced to Newton, says that the frictional force is proportional to the motion of the probe along the substrate and that it opposes that motion. We do not view this as the “true” equation of friction but rather as a plausible hypothesis that is to be tested by comparing the predicted behavior against measurements. The model is unphysical in that it accords the same frictional force for a given horizontal velocity for any surface separation or load. In practice however this does not appear to be an important consideration because the only time that there is lateral motion of the probe on the surface is when the surfaces interact. Obviously one expects a frictional force whether the normal load is positive or negative, and the model encapsulates this idea. The next order analytic model would be one that included a term quadratic in the normal load, $F_y = -(\xi + \xi F_z^2)y_2$. The present expression may be viewed as the leading term in an expansion about zero load. We calculate the dynamic trajectory of the surfaces in typical AFM measurements, and we conclude not only that can friction cause hysteresis in the constant compliance regime but also that friction can effect dynamic surface force measurement in the AFM prior to contact.

Figure 5 shows the angle of deflection as a function of position for the driving velocity of $z = \mu \text{ms}^{-1}$. The friction coefficient used here, $\xi = 0.1 \, \text{kg s}^{-1}$, gives for this driving velocity a friction force $F_y \approx 3 \, \text{nN}$ in the constant compliance regime. Hence the results should be compa-
AFM, the jump (signified by constant while the outward run proceeds past the point where they with the steady state analysis. Finally, there is an run (unloading) lies above the inward run, in agreement pure normal mode. As already mentioned, the outward in part an artifact of the coarse grid used for plotting and the cantilever. The irregular nature of the vibrations are the trajectory, mainly due to the induced vibrations in without a very high sampling rate.

for less than 0.1 ms), and in practice it may not be visible this case in total less than 1.5 ms, with it being positive for less than 0.1 ms, and in practice it may not be visible without a very high sampling rate.

As pointed out in I, one has to distinguish between the actual deflection of the cantilever and the apparent deflection deduced from whatever parameter that one is actually measuring. In the case of the AFM one measures the deflection angle, and one deduces the apparent deflection $\xi(\theta) = \beta \theta$. The deflection when $\theta > 0$, even though the actual deflection has $x < 0$. The magnitude of the spike obviously depends on the friction force and also on the parameters used to characterize the AFM. Hence the magnitude of the spike will be rather sensitive to the model used. It is of extremely short duration (in this case in total less than 1.5 ms, with it being positive for less than 0.1 ms), and in practice it may not be visible without a very high sampling rate.

After contact there is a noticeable increase in noise in the trajectory, mainly due to the induced vibrations in the cantilever. The irregular nature of the vibrations are in part an artifact of the coarse grid used for plotting and in part a result of the fact that the cantilever is not in a pure normal mode. As already mentioned, the outward run (unloading) lies above the inward run, in agreement with the steady state analysis. Finally, there is an additional hysteresis as the surfaces remain in contact while the outward run proceeds past the point where they originally jump into contact. In this case one would have to drive out to about $z = -|F_z(z_0)/k| = -200$ nm before the surfaces jumped out of contact. This particular difference between the inward and outward runs is not due to friction but is the well-known result of the adhesion between the bodies and it is often used to measure their surface energy.

Figure 5 compares calculated deflections for various driving velocities and friction coefficients with the deflection expected from a static measurement (i.e., Hooke's law, $F_z = kz_2$). Although the dissipative drainage term has been included in the calculations, it has been checked that it had no significant effect. It was also checked that inertial effects were similarly negligible. Hence the velocity dependence apparent in the figures arises solely from friction. This is perhaps a surprising finding: friction causes the magnitude of the surface force to be underestimated. In part this is a consequence of the model that was invoked for friction, namely, $F_y = -\xi y_2$. But as argued above, there is every reason to believe that friction operates whenever there is an interaction between the surfaces, and this includes the precontact regime that is exhibited in the figures. The movement of the probe horizontally across the surface during the jump into contact (due to the inclination of the cantilever) is greatly reduced by friction. Hence there is an effective spring constant that is higher than the force measuring spring constant because the cantilever cannot deflect in its preferred manner.

In fact, during the jump into contact the deflection decreases (becomes more negative) while the angular deflection increases (becomes more positive), in order that $y < 0$. This is the spike discussed in Figure 5, which is seen in each of the dashed curves in the present figures. This is the apparent deflection introduced in I. In the AFM the angle $\theta$ is measured (by the light lever) and the deflection is deduced via the calibration factor $\beta$ that is measured in the constant compliance regime. It is clear from Figure 5 that $\beta$ is well-defined and in this case is found numerically to equal 1.3 x 10$^{-2}$ rad$^{-1}$, which agrees with that calculated from the analytic result, eq 15. By use of the calibration factor, the apparent deflection and the apparent separation are

$$\dot{\xi}(\theta) = \beta \theta \quad \text{and} \quad \ddot{\xi}(\theta) = \ddot{\xi}(\theta) - z(t) \quad (29)$$

![Figure 5](image1.png)

**Figure 5.** Calculated trajectory for $z = 0.1 \mu m s^{-1}$, for the van der Waals interaction ($A = 5 \times 10^{-15}$, $z_0 = 0.35$ nm, and $R = 100$ nm), in the presence of friction, $F_y = -\xi y_2$, $\xi = 0.1$ kg s$^{-1}$, cantilever parameters as in Figure 3. The hysteresis in the constant compliance regime is clear in the smoothed data in the magnified inset, where the outward run lies above the inward run.

![Figure 6](image2.png)

**Figure 6.** Actual ($z_0$, full curve) and apparent ($\dot{z}_0(\theta)$, dashed) deflections as a function of actual or apparent separation for the van der Waals interaction for an AFM cantilever (parameters as in the preceding figures). The upper pair of curves is for a driving velocity of $z = 1 \mu m s^{-1}$, and the lower pair is for $z = 0.1 \mu m s^{-1}$. The bold curve is the deflection that would occur in a static measurement (shown dashed in the unstable region). The friction is $F_y = -\xi y_2$. (A) $\xi = 0.01$ kg s$^{-1}$. (B) $\xi = 0.1$ kg s$^{-1}$. (C) $\xi = 0.1$ kg s$^{-1}$.
It can be seen in the figures that prior to the spike there is reasonable agreement between the apparent and the actual deflection. After contact one sees that there is an apparent shift in the zero of separation. This in essence arises from the constant frictional force in the steady state constant compliance regime. The shift is proportional to the driving velocity and to the friction coefficient. Experimentally the only way to detect this shift is from the hysteresis on the inward and outward runs. Ignoring the hysteresis, the zero of separation is commonly established experimentally by moving the constant compliance regime on the inward run so that it lies at zero separation. But the effect of this shift is a artificially increase the magnitude of a measured deflection at a given position. That is, the underestimated force measured at a given separation is attributed to a larger separation, where it would be weaker anyway. Hence one does not have a quantitative measurement of the van der Waals attraction, but rather the appearance of one that results from the partial compensation of two artifacts: the precontact diminished deflection and the shift in the postcontact zero of separation.

3. Experimental Section

Atomic force microscopy was performed with an AutoProbe with contact mode head (Park Scientific Instruments, Sunnyvale, CA). The lower surface was a flat glass plate, cleaned prior to use in ethanol and mounted on a Scanmaster piezo-driver, the movement of which was monitored by the attached position sensitive photodetector. The upper surface was the probe of a triangular silicon ultralow of nominal spring constant of 0.26 Nm⁻¹ (Park Scientific Instruments, Sunnyvale, CA), the deflection of which was monitored from the movement of the reflected laser beam across the face of the photodiode. The vertical split. The movement of the piezo-driver was calibrated interferometrically, and the cantilever spring constant was calibrated gravitationally. All measurements were performed in air open to the atmosphere and at room temperature (22.7 °C).

This section is divided into two parts. First a characterization of the AFM is given. It turns out that hysteresis due to friction is rather small, and it can be masked by certain experimental artifacts. An accurate and reliable measurement of distance is required, and here we test the two methods commonly used in the AFM (the nominal driving distance of the piezo-motor and the distance measured with an attached position-sensitive detector) against an absolute interferometric measurement of distance. The second part gives AFM measurements of the van der Waals attraction and discusses the evidence for friction in these results.

3.1. Characterization. Atomic force microscopes commonly measure the cantilever deflection by shining a laser on to its end and following the movement of the reflected beam across a split photodiode. There can also be substantial reflection from the substrate, and due to the coherence of the laser this interferes with the primary beam at the photodiode. The path difference, which determines whether the interference is constructive or destructive, is linearly proportional to the vertical distance between the substrate and the end of the cantilever. Changes in this distance are manifest as changes in the intensity measured by the photodiode. The measured variation of intensity with distance in the large separation, zero force regime is not due to changing deflection of the cantilever but rather to changing path difference, and the consequent interferogram provides an absolute measure of distance changes. This can be used to characterize the device and to characterize the performance of the various experiments. To be specific, the signal intensity is given by

\[ I(z) = I_0 \cos(\Delta z / \lambda) \]

where \( \Delta \) is a geometric factor that depends on the wavelength and angle of the laser beam.

Figure 7 uses various means of distance to display a single force measurement of van der Waals attraction in air. The measurement commences with the surfaces in contact, first driving out beyond the point where the surfaces jump apart and then immediately reversing for the inward run. In Figure 7A the nominal position of the piezo-motor is used as the measure of distance. That is, the known voltage applied to the piezocrystal is multiplied by a calibration constant to obtain the distance. (This is the most common method used in AFM measurements.) The problem with this method is that piezoelectric materials are only linear for small changes. As the applied voltage and distance are increased, the nonlinear behavior becomes noticeable, and there is hysteresis between the inward and outward runs. The magnitude of the effect depends on the particular piezocrystal and varies with the applied voltage and the driving velocity. The nonlinearity and hysteresis are most apparent from the interference fringes in the inset of Figure 7A. The fringes on each run are not uniformly spaced, which indicates nonlinearity. Further, the inward run is not superimposed on the outward run, which indicates hysteresis. One concludes that the voltage applied to the piezo-motor does not give the position of the substrate with acceptable accuracy for typical force measurements.

In the constant-compliance regime, one sees that using the nominal position shifts the inward run to larger distances than the outward run. By coincidence this is qualitatively what one would expect if friction were operative, as was shown in Figures 3–6. However, the ≈1.5 µm shift in Figure 7A is entirely an artifact of using the nominal piezo-motor position as the distance scale. In the inset it can be seen that the interference fringes on the inward run are shifted by about the same amount with respect to the outward run. The figure demonstrates that the nominal position moved by the piezo-motor cannot be used to measure hysteresis due to friction.

This conclusion is significant in view of the study of contact hysteresis by Hoh and Engel. In that study the nominal position of the surface was used, and therefore the reported shifts in the contact lines (25–75 nm, in air) appear due at least in part to piezo hysteresis, and friction may not be the major contributor to the measured phenomenon. It is difficult to use the present results to quantify precisely the effect of the friction in that study because of the difference in piezoscanners and driving velocities. It could well be smaller than that found here if smaller driving distances than here were used. Although our theoretical analysis above supports the qualitative conclusions about the effects of friction that were drawn by Hoh and Engel, the results in Figure 7A suggest that quantitative measurements of contact line hysteresis cannot be made using the nominal position of the piezo-motor without characterizing the hysteresis in that motor.

It is emphasized that piezo hysteresis is a commonly occurring phenomenon and that all atomic force microscopes are affected to greater or lesser degree, depending upon the particular device specifications. High-precision work demands that this artifact be characterized in each case, such as in Figure 7A.

uniformly spaced, which demonstrates that the device is attached to the housing. The interference fringes give its motion with respect to light-emitting diodes photodetector attached to the driving piezo-scanner, which is responsible for much of the noise in the postcontact force-separation curves. Increasing the shift would cause the outward run to lie at larger, more positive positions than the inward run in the constant compliance regime. It is difficult to think of a physical mechanism that would cause this behavior. Increasing the shift would displace the two constant-compliance curves in a manner expected from friction. From the fringes we estimate the upper limit on the amount of hysteresis displacement due to friction in this system to be about 10–15 nm. Alternatively, the fact that overlaying the constant-compliance lines also approximately superimposes the interference fringes is consistent with the hypothesis that friction between the tip and the substrate is negligible in this example.

The apparently constant displacement of the interference fringes on the inward and outward runs has the appearance of backlash. We are not entirely sure of its origin. It is possible that the photodetector attached to the piezo-tube rubs against the light-emitting diodes attached to the housing and that one or other of both are displaced by about 20 nm on the inward and outward runs, depending upon the velocity.

Figure 7C we have shifted the inward run so that it coincided with the outward run in the constant-compliance regime. The amount of shift that was required to overlay the two curves was +57 nm. From the inset it can be seen that the interference fringes are now largely superimposed on the runs. Obviously due to the noise in the interference pattern there is some uncertainty in the degree of fit. We estimate that varying the shift by about ±10 nm would still give a reasonable coincidence of the fringes. Decreasing the shift would cause the outward run to lie at larger, more positive positions than the inward run in the constant compliance regime. It is difficult to think of a physical mechanism that would cause this behavior. Increasing the shift would displace the two constant-compliance curves in a manner expected from friction. From the fringes we estimate the upper limit on the amount of hysteresis displacement due to friction in this system to be about 10–15 nm. Alternatively, the fact that overlaying the constant-compliance lines also approximately superimposes the interference fringes is consistent with the hypothesis that friction between the tip and the substrate is negligible in this example.

The constant-compliance curves are noticeably smoother in Figure 7C than in parts A and B of Figure 7. This is because we have here applied an 11-point running average to the z-detector position, to correct for a digitization error in the photodetector. The procedure is equivalent to linear interpolation between successive digitization levels, which is valid because the motion of the driving piezo is locally linear. The digitization error in the photodetector is most apparent at high sampling frequencies, and it is generally responsible for much of the noise in the constant-compliance-separation curves.

These two artifacts of the position-sensitive detector—backlash and digitization—are peculiar to the Park Instrument, whereas as mentioned above piezo hysteresis occurs generally in all atomic force microscopes. On balance the position-sensitive detector performs well and it makes the device suitable for high-precision force measurements, despite the backlash precluding accurate measurements of friction.
compliance regimes scales roughly with the driving speed. In Figure 8A, $z = 6.6 \mu m s^{-1}$ and the shift is $+27$ nm. This is similar to the case $z = 8.4 \mu m s^{-1}$, where the shift was $+33$ nm (not shown). In both cases the interference fringes are reasonably superimposed by this shift, to within about $\pm 10$ nm. For a slow driving velocity of $z = 0.4 \mu m s^{-1}$ (Figure 8B), no shift appeared to be necessary as the constant-compliance lines were already in relatively good coincidence, as were the fringes. (We drove over a quite small distance in this case in order to increase the sampling frequency.) These shifts can be compared with the $+57$ nm shift of Figure 7C, for a driving velocity of $z = 17 \mu m s^{-1}$. That the shift increases with increasing velocity tends to support the above interpretation that the backlash is due to rubbing between the photodetector and emitter.

The results in Figures 7 and 8 set an upper limit of about $10$ nm for the amount of hysteretic displacement due to friction, and they are not inconsistent with the possibility that friction is negligible in this system.

3.2. van der Waals Attraction. Figure 9 shows the measured van der Waals attraction between a silicon ultralever and a glass substrate for several driving velocities. The inward run and part of the outward run are shown in each case and compared to the static deflection expected from van der Waals theory. Note however that the variability in the measured jump-out distance, $200 \pm 40$ nm, makes the precise values of the magnitude of the deflection somewhat uncertain. To convert these deflections (nm) to force (nN), multiply by $k_0 \cos \beta_0 = 0.21 N m^{-1}$. To convert force to the interaction free energy per unit area between planes, divide by $\pi R$, with $R = 220$ nm.

An additional cause of uncertainty in the comparison of the experiments with each other and with theory is the choice of the absolute separation. As discussed above there appears to be backlash in the separation measured by the position-sensitive detector in the AFM, and so in the figures we have shifted the inward run so that the constant-compliance line overlaps with the outward run in that regime. Further, we have chosen the zero of separation so that these constant-compliance lines lie at a separation $h = z_0 = 0.35$ nm. The bold curve is the theoretical van der Waals attraction (static, shown dashed in the unstable region), for a Hamaker constant of $A = 5 \times 10^{-19}$ J, a spring constant of $0.21 N m^{-1}$, and an effective radius of $R = 220$ nm, as derived from the measured jump-out distance of $200$ nm. The circles represent the calculated dynamic trajectory for a mass of $1 \mu g$, sampled at the same rate as the respective experiments. (A) The driving velocity is $z = 0.82 \mu m s^{-1}$, the sampling frequency is $1.6$ kHz, and the shift is $+2$ nm. (B) $z = 1.36 \mu m s^{-1}$, $2.7$ kHz, and $+4$ nm. (C) $z = 2.0 \mu m s^{-1}$, $4$ kHz, and $+7$ nm.
We used a mass of 1 g, and the measured deflection is quite unambiguous. In this case the maximum variation in the magnitude, the functional form of the measured data is as expected, and it can be concluded that this indeed a true measurement of the van der Waals force. The evidence at the lowest driving velocity (0.82 μm s\(^{-1}\), Figure 9A) is particularly convincing, since the experimental data follow the static van der Waals theory up until the jump into contact. The two faster runs (1.4 and 2.0 μm s\(^{-1}\), parts B and C of Figure 9, respectively), begin to underestimate the van der Waals attraction prior to the jump into contact. The high sampling rate gives a sufficient number of data points in the interaction region to conclude that the discrepancy is significant.

Figure 10 shows the precontact data on an expanded scale and includes an additional fast run (7.2 μm s\(^{-1}\), sampled at 1.6 kHz) and a repeat experiment of the slowest run (sampled at 800 Hz). The quantitative agreement between the duplicate runs gives one some confidence in the data. The underestimation of the precontact attraction is quite evident, and it appears to increase systematically with increased driving velocity. Despite the low sampling spatial frequency, the discrepancy between the predicted and the measured van der Waals attraction for the fastest run is quite unambiguous. In this case the maximum measured deflection is −1 nm, which is significantly less than the −6 nm of the other cases. Figure 10 confirms that there is a significant underestimate of the precontact attraction in the measured data and that this is a consequence of the dynamics of the measuring process.

Before ascribing the dynamic effect apparent in the data to friction, we need to eliminate the possibility of inertial or aerodynamic drainage effects. We tested these latter by performing dynamical calculations for the simple spring model of the AFM. We used a mass of 1 μg, which is about a thousand times larger than the mass of the bare cantilever, giving an upper bound on inertial effects. For the drainage term we used the viscosity of air, 1.8 × 10\(^{-5}\) kg m\(^{-1}\)s\(^{-1}\). As can be seen in Figure 9, the results of these dynamic calculations are in good agreement with the static calculations for all the experimental driving velocities. We conclude that neither inertia nor viscosity is responsible for the underestimate of the precontact attraction.

The second piece of evidence for precontact friction is the distance from which the surfaces jump into contact. From the static calculations, the gradient of the van der Waals attraction first exceeds the spring constant at h = 4.4 nm. The sampling points for the dynamic calculations show that there is a relatively large acceleration of the surfaces at this point; the spacing of the sampling points gives a good indication of the jump position. In contrast the experimental data exhibit the jump from significantly larger separations than this. The actual jump position is too large by at least 1 nm (Figure 9A), 3 nm (Figure 9B), and 2 nm (Figure 9C). This suggests that the curves should be shifted to somewhat smaller separations. Such a shift would make the underestimate of the precontact attraction even more marked. Also, it would cause the apparent hard-wall contact to lie at negative separations, and it would cause the outward constant compliance line to lie at more positive positions than the inward one. These three effects are predicted by the friction calculations in Figure 6, and so such a shift would certainly be consistent with the notion that friction is operative in this system.

Because we do not know the value of the friction coefficient, we cannot quantitatively show that it is friction that is responsible for the departures from the van der Waals theory. The interference fringes do not have the resolution to establish definitively the relative shifts and hysteresis in the inward and outward runs. There could be other possible explanations for the acceleration and jump so far from contact. In particular, we were unable to control the humidity of the atmosphere in these experiments, and we cannot rule out the possibility of capillary condensation contributing to the measured forces. This might occur rapidly enough on approach to contact, and either the shift in the plane of origin of the force due to wetting layers on the surfaces or bridging between the two surfaces would lead to a larger than van der Waals attraction causing a premature and more rapid jump into contact. It is not inconceivable that the time scale for such putative condensation is comparable to the time during which the surfaces interact and that the decreased attraction at higher driving velocities may also be a consequence of capillarity.

There is one further noteworthy feature in the experimental data. Postcontact one can discern vibrations or oscillations in the curves. These are particularly clear in parts B and C of Figure 9, where they can be seen in both the inward and outward runs. (We have seen similar vibration in other, unpublished, data.) The frequency of the vibrations in Figure 9B is about 640 Hz, and that in Figure 9C it is about 670 Hz. The fact that the two different driving velocities give the same temporal frequency but different spatial frequencies suggests that the phenomenon is not due to corrugations or other surface features. On the other hand, the fact that the oscillations on the inward and outward runs are in phase does point to such a possibility, or to stick-slip motion along the surface. This appears the most likely explanation of the phenomenon. Although this postcontact behavior is reminiscent of that seen in I, where it was ascribed to cantilever oscillations and to elastic vibrations, these can be ruled out in this case where they would be of the order of 40 kHz.

4. Conclusion

This paper has mainly been concerned with the effect of lateral friction on the measurement of normal forces with the atomic force microscope. Two features of the instrument motivated this investigation: first the automated nature of the experimental protocol, and second the use of an inclined cantilever. The computerized control and data acquisition system mean that the AFM technique ought to be classified as dynamic surface force measurement, and we had demonstrated in the first paper in the
series\(^1\) that dynamic effects were significant in other computer-controlled devices. Here it is shown, however, that in the AFM inertia was generally not an important consideration unless a large colloidal probe was used. The second feature that we analyzed was the effect of the angle of inclination of the cantilever. A relatively trivial consequence of this is that the cantilever spring constant needed to be corrected by a trigonometric factor to obtain the force measuring spring constant. A more important consequence of the inclination is that normal motion of the substrate causes lateral motion of the tip along the substrate. It is the frictional force induced by this motion that effects the measured surface force.

Three effects were identified from model friction calculations: hysteresis in the constant compliance lines when the surfaces were in contact, a shift in the apparent position of the surfaces, and an underestimate of the magnitude of the precontact surface force. The first two of these effects have been discussed previously,\(^{12,14}\) and the present analysis enabled a quantitative calculation to be made for a variety of models. Figure 11 shows how friction can cause the AFM tip to appear to be vertically shifted and, on an inward run, to appear to have penetrated the sample. Since friction would operate in the opposite direction on an outward run, the two constant-compliance lines will appear displaced. It is this displacement of the constant-compliance lines on the inward and outward runs that was shown to be the primary signature of friction.

Such a hysteretic displacement is very common in AFM data, and indeed it had previously been used as a quantitative measurement of friction.\(^{14}\) However the experimental data presented here showed that the hysteresis in the piezoelectric crystal used as a driving motor causes exactly the same displacement. By using interferometry, we were able to show that the hysteresis in the present measurements was almost entirely due to using the nominal position of the surfaces (i.e., it was due to the nonlinearity and hysteresis of the piezocrystal itself). In contrast, using the actual position of the surfaces, as established by interferometry, within the accuracy of the interferometry we could not detect any hysteretic displacement of the constant-compliance lines. This placed an upper limit on the magnitude of the friction force that could be operative in this system, and it casts some doubts on the quantitative results of Hoh and Engel.\(^{14}\)

We used the AFM to measure the van der Waals force between solid surfaces in air. We obtained a significant precontact attraction, and the functional form was as predicted by theory. This was most evident at low driving velocities. As the driving velocity increased, the magnitude of the measured force decreased, in qualitative agreement with the prediction made on the basis of the model friction calculations. However we could not definitively attribute it to friction because of the possibility that capillary condensation was contributing to the phenomenon. In either case, the unambiguous dynamic effect that we saw is sufficiently interesting to warrant further investigation.

The novel prediction that friction could cause a diminution of the precontact force was made on the basis of theoretical calculations using a simple model of friction, namely, that it was proportional to the lateral velocity of the tip across the substrate. We argued that friction will occur between mutually sliding and interacting bodies and that this includes the precontact interaction. However it would be true to say that the functional form of the model friction force, and in particular whether it should include some proportionality to load and to separation, is a topic for further work. We point out one situation where such precontact friction is known to occur, namely, for two shearing surfaces in a liquid. In this case the frictional force is proportional to the sliding velocity, the shear viscosity of the liquid, and the reciprocal of the separation. Given this fact, the calculations presented in this paper indicate that surface forces will in general be underestimated by the AFM in liquids. A quantitative study of the effects of inertia, drainage, and shear viscosity on dynamic surface force measurements in liquids will be presented in future work.\(^{33}\)

(33) Attard, P. In preparation.

Figure 11. An AFM tip at contact, shown in the absence of surface forces by the dotted line and the light-shaded stylus. For an upward moving substrate (inward run), a friction force to the right acts on the stylus (black), which exerts a torque on the cantilever causing an s-bend (bold line). Measuring only the angle of the tip of the cantilever would give the appearance that the substrate had been penetrated (dark shading). Friction is in the opposite direction for an outward run, reversing the angular deflection, which causes the two constant-compliance curves to appear displaced.