Near-field scanning optical microscopy (NSOM) uses shear-force feedback as the primary method to control the probe-sample distance. We describe the nonlinear interaction between the tip and sample with a simple truncated driven harmonic oscillator model. The model accurately describes the measured dynamics of this system. Insights are gained into the mechanism behind this interaction, and we give strong evidence that the probe taps on sample surface adlayers in normal operation, but will tap the underlying sample surface when the oscillation is nearly quenched.
Distance regulation in near-field optical microscopy (NSOM) makes use of the force interaction between the probe and sample. There are several experiments in which accurate distance control is essential: fluorescence lifetime [1], gradient field Raman [2], and electromigration [3,4]. Understanding the dynamics of the probe interaction and the mechanism behind the distance control helps to optimize distance regulation and evaluate its accuracy. We find that a nonlinear interaction such as a tapping force is required to accurately describe the resonance behavior of the probe both in and out of feedback (near and far from the sample). Furthermore, our novel study of the system dynamics shows that the model is able to predict the detailed nature of the time response of the probe that we observe in our measurements. Finally, we present strong evidence indicating that the probe is tapping on surface adlayers prior to tapping on the surface itself as the probe approaches the sample, and we comment on the suitability of shear-force feedback for accurate distance control.

Shear force feedback using a tuning fork oscillator [5] is one of the most widely used techniques for controlling tip sample separation with an NSOM. It relies on voltage generation by a crystal tuning fork to measure the oscillation amplitude that varies with tip sample distance. The oscillation amplitude decreases when the probe is close to the surface due to an increase in damping and a shift in the resonance frequency. A variety of mechanisms and combinations of mechanisms are proposed to be responsible for the interaction between the probe and the sample such as friction [6], tapping (or knocking) [7-9], distance dependent probe bending [10], damping layer [10-14], and coulomb fields [15]. In this paper we show evidence of a nonlinear tip-sample interaction. Of these mechanisms, only tapping and probe bending have a nonlinear response. Figure 1 (a) shows the resonance curve far from the surface and its evolution as the
probe is moved closer and closer to the surface. The resonance frequency shift we see implies that a nonlinear mechanism is active. The obvious nonlinear interaction is tapping.

To model this tapping interaction, we use a simple truncated driven harmonic oscillator used by others to model NSOM [7] and atomic force microscope [16] probe-sample interactions. In this model the tapping force is included in the addition of a strong force when the lateral position of the tip exceeds a critical value, $x_c$. The equation that describes such a system with effective mass $m_{\text{eff}}$ driven by a force $F_{\text{drive}}$ is

$$F_{\text{drive}}/m_{\text{eff}} = F_0 \cos(\omega_d t)/m_{\text{eff}} = \ddot{x} + 2\beta_0 \dot{x} + \omega_0^2 x + H(x-x_c)[\omega_1^2 (x-x_c) + 2\beta_1 \dot{x}],$$  \hspace{1cm} (1)

where $\omega_d$ is the radial tip oscillation driving frequency, $\beta_0$ and $\beta_1$ refer to damping and $\omega_0$ and $\omega_1$ the resonance frequencies of the fork/fiber oscillator and fiber/adlayer system, respectively, and $H$ is the step function. We find $\omega_0$ and $\beta_0$ from the free resonance curve. These parameters depend only on the tuning fork/probe assembly. From a combination of the free resonance curve ($H=0$) and a shifted, damped resonance curve we determine $\omega_1$ and $\beta_1$. We comment in more detail on the significance of the values of these two parameters for the physics of the tip-sample interaction later in the paper. The parameter $F_0/m_{\text{eff}}$ is used to normalize the equations so that the amplitude of $x$ is 1 with no tapping ($H=0$). Numerical solutions to the model for various values of $x_c$ give resonance curves as a function of the fraction of the undamped resonance peak (setpoint) that are consistent with the resonance curves we find in our experiments (see figure 1). Note that in both the numerical analysis and in the experiment, the damped resonance curve crosses over the free resonance curve on the high frequency side resulting in driving frequencies unavailable for normal feedback operation without switching the polarity of the feedback response.
We study the dynamics of the probe sample interaction by observing the calculated oscillation $x(t)$ as we add or remove the step function terms from the model. When we add the step function, we refer to the system as tapping on, and when we remove the step function, we refer to the system as tapping off. This is analogous to the probe being moved from a position out of feedback to a position close to the sample for the tapping on and vice versa for the tapping off. Figure 2 (a) shows the time response of the model overlaid with the resonance curve. The time response becomes faster as we move off resonance in either direction, and the time response for turning the tapping off approaches the time response for turning the tapping on. At the resonance frequency, the time response for the tapping on vs. off is very different.

The dynamic behavior of the probe predicted by the model is consistent with what we would expect qualitatively. Recall that there are two parameters responsible for the time response of the system when it is in feedback – the change in damping and the frequency shift – both can be seen in figure 1. The time response is limited by the width of the resonance curve, in the sense that if the tip gets too far from the sample, the decay time for the oscillation scales inversely with the peak bandwidth. Thus the time response is slow due to the high quality factor (100 to 700) combined with the relatively low resonant frequency (32-40kHz). When the probe is close to the sample, the oscillation decreases due to both the damping of the probe and the frequency shift, and while the damping is slow, the peak shift is fast. Near resonance, at probe positions relatively far from the sample, the slow damping is dominant; at probe positions close to the sample the fast peak shift is dominant. This inequity results in the two different curves for the cases of tapping on and tapping off. When the probe is off resonance, the peak shift is dominant, and the time response is faster for both cases.
In our experiments we clearly see the dynamic behavior predicted by the tapping model. Without lateral scanning, we apply a 30 nm trapezoidal shaped pulse with an 8 msec rise and fall time to the z-piezo, which ramps the tip alternately towards and away from the surface. The probe begins in feedback, so when it moves towards the surface, the tapping is increased (tapping on), and when it is pulled away from the surface, the tapping is decreased (tapping off). We maximize the gain until there is no overshoot when the probe retracts as a reaction to being pushed towards the surface. We plot the time response for the inward motion of the probe as a reaction to being pulled away from the surface as a function of driving frequency, and we see a behavior that is very similar to that predicted by the model, figure 2b.

In a 2nd experiment to study the time response of the system, we apply a square wave to the z-piezo that moves the tip 40 nm alternately towards and away from the surface, while monitoring the system response. The feedback gain is the same for all frequencies. We expect the time for the system to respond when the pulse pushes the probe towards surface to be fast in all cases, pulling the probe away from the surface, due to the increased tapping. Since the gain is high, overshoot is observed when the ‘ingoing’ response is too slow. This will be the case without strong nonlinear effects (at frequencies near the resonance peak). Figure 3 shows exactly this behavior. The curves shown are the in and out motion of the probe with the resonance curve overlaid for reference. As the pulse pulls the probe away from the surface, the time response for the inward motion, inversely proportional to the slope, decreases as we increase the driving frequency above the resonance peak until the time response for the in and out motion of the probe align. This balanced response quenches the overshoot. If this frequency rather than the peak frequency were used for feedback, the bandwidth of the NSOM could be
increased [17]. Further increases in frequency much beyond this point result in instability since the damped resonance curve begins to cross over the free resonance curve.

With confidence in a model that accurately describes both the static and dynamic aspects of our system, we look more closely at the choice for $\omega_1$ and $\beta_1$. Gregor uses the clamped resonance curve ($x_c = 0$) to determine these values, and they accurately describe the large frequency shifts and minimal additional damping of his resonance curves as the probe approaches the sample. Figure 4 shows several of our resonance curves including the clamped curve. The trends are qualitatively different from Gregor's observation. We find that using the clamped curve to determine $\omega_1$ provides values that give a poor fit to our data because the spring constant is too large. The frequency shift for the clamped peak is 400 Hz as compared to 60 Hz for the damped peak we use. A simple picture describes both our and Gregor's observations: for most of the approach curve, we are tapping on (soft) adlayers compared to Gregor who is tapping on the sample surface or hard, frozen (low temperature NSOM) layers. Therefore, we must obtain the parameters for our model from the adsorbed layer tapping, not surface tapping if we are to describe the dynamics under normal ambient operation. We note that rather than tapping on the surface adlayers, the probe may be opening a cavitation hole within the adlayers, resulting in a tapping on the sidewalls of this cavitation hole. Near a hydrophilic surface, water is structured [18] as are other near-surface solvents [19], so they are less mobile, indicating that cavitation is likely with the ultrasonic tip oscillation frequency. Cavitation is also possible with less mobile adsorbates and the less structured water (clathrates) near a hydrophobic surface [20]. Either case, tapping on the adlayers or cavitating in them, is consistent with our observation of the change in stiffness that we observe when the probe reaches the sample surface.
If the probe is tapping on the sample surface, then the distance to the sample depends both on the fiber oscillation amplitude and the angle between the sample and the probe [9]. However, if the probe is tapping on surface adlayers, and the oscillation amplitude of the probe is much less than the thickness of the adlayer, then the distance to the sample depends primarily on the thickness of the adlayer and can be accurately controlled, as is the case with our system. In our studies, the tip does not tap on the hard sample surface (beneath the adlayers) in the normal operating range, or even closer to the surface, until the probe oscillation amplitude has decreased due to sample interaction to a few percent of the free oscillation value. This is clear from the small measured oscillation amplitude at the feedback frequency (~1 nm) [17] compared to the 5-10 nm approach curve – the tip simply cannot reach the surface. As an additional evidence, in our electromigration studies [3,4] we have found that we cannot measure tunnel current between the tip and sample unless we set the feedback level much below the normal operating range (probe very close to the surface). The process is repeatable, with minimal tip wear determined by little change in resolution even after several hours at a low feedback level (tunneling). In the case of small probe oscillation amplitudes on surfaces with a contamination layer (typical case), shear-force is an accurate measure of probe-sample separation.

In conclusion, the nonlinear model accurately describes both the system dynamics and the resonance curve behavior as the probe approaches the sample. During this approach the probe taps on surface adsorbed layers prior to tapping on the surface itself. This implies that the lateral force feedback is a good indicator of tip-sample distance when small oscillation amplitudes are used, and that a tapping mechanism describes the nonlinearity of the tip-sample interaction. This nonlinear interaction can be used to increase the bandwidth of the high Q tuning-fork-based distance regulation system.
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References:


**Figure 1:** a) Typical resonance curves obtained with the tuning fork method of oscillation amplitude measurement at various tip-sample separations (solid lines). b) Resonance curves with different degrees of tapping obtained by a numerical solution of the nonlinear differential equation (dashed lines).

**Figure 2:** a) The numerical calculation of the time response for the feedback signal to drop to 1/e for a variety of driving frequencies for two situations: turning the tapping off and turning the tapping on. This time response is overlaid with the resonance curve for reference. b) The experimental time response for the probe to find the surface with optimized gain given an 8msec-ramped trapezoidal step of height 30 nm.

**Figure 3:** The feedback response to an impulse of 40 nm on the z-piezo for a variety of driving frequencies on either side of the resonant frequency with constant gain. The black lines are the outward motion. The grey lines are the inward motion. The resonance curve is overlaid for reference.

**Figure 4:** Experimental resonance curves showing the undamped resonance, a resonance at 35% of the free resonance and a clamped resonance curve. The peak of the clamped resonance is only 2% of the undamped resonance peak, and the frequency shift is ~400Hz.
Figure 1 (C. L. Jahncke, S. H. Huerth, Beverly Clark III and H. D. Hallen)
Figure 2a and b (C. L. Jahncke, S. H. Huerth, Beverly Clark III and H. D. Hallen)
Figure 3 (C. L. Jahncke, S. H. Huerth, Beverly Clark III and H. D. Hallen)
Figure 4 (C. L. Jahncke, S. H. Huerth, Beverly Clark III and H. D. Hallen)