The Physics of Phase Imaging:
Attractive vs Repulsive Mode Imaging

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Background

For several years in AFM, phase imaging (mapping the phase of the oscillating cantilever relative to the drive in AC modes) was a "looks great but what does it mean?" signal. Unlike many of the "new" signals in AFM which so often look like a derivative of topography, it was clear that the phase images were truly different especially on polymer systems, but nobody understood the contrast mechanism. Different publications attributed the contrast to stiffness, viscoelasticity, surface energy, etc. Based on work in the last few years[1,2], it is clear now that the phase signal is directly related to two things: (1) Whether the cantilever is operating in net attractive or repulsive mode, and (2) The energy dissipated by the tip-sample interaction. All of the earlier findings can be understood in this framework. For example, total energy dissipation in a polymer does depend on the stiffness, because the stiffness determines how much the polymer deforms and therefore how much energy is lost to internal friction. A polymer sample with an infinite storage modulus will not deform at all and therefore won't dissipate any energy even if it has a large loss modulus.

Just because we understand that phase is energy dissipation doesn't mean we're done understanding phase, but that we are just getting started. Imagine that early in the field we didn't know that the bending of the cantilever was related to force and we had imaged using this mystery signal "Deflection". Understanding that deflection was directly related to the force on the cantilever would be the equivalent breakthrough. You only measure one quantity (the force on the cantilever in the z direction) but there are many contributors. The force can have contributions from wetting, Van Der Waals, electrical double-layer, and many other forces. We are always stuck with more unknowns than measured quantities. However, experiments can root out the causes. For example, adding salt to a solution and seeing a big change in the force can implicate double-layer forces. Eventually you can just look at a force curve and have a good guess as to what is causing the force: you learn the fingerprints for different forces. Similarly, sources of energy dissipation can be tracked down and learned as well.
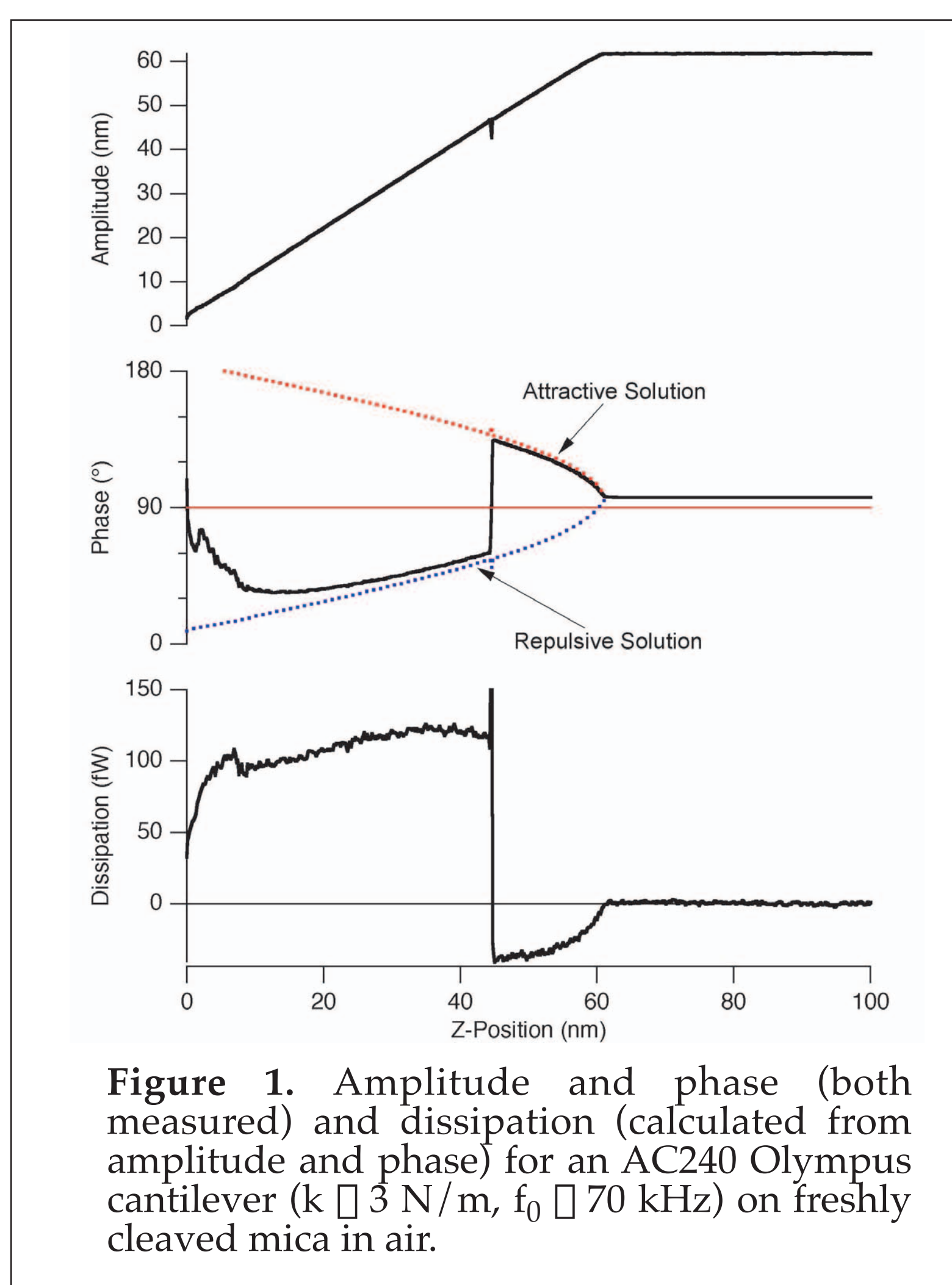

Figure 1. Amplitude and phase (both measured) and dissipation (calculated from amplitude and phase) for an AC240 Olympus cantilever (k ≈ 3 N/m, f_0 ≈ 70 kHz) on freshly cleaved mica in air.

Attractive and repulsive solutions coexist and have very different energy dissipation

One of the striking things to come out of the physics of these AC modes is the coexistence of two solutions. For a fixed experimental situation (cantilever amplitude, setpoint, etc.), there are really two types of behavior possible. The easy way to distinguish these two solutions is to look at the phase. One solution will have the phase greater than 90. This solution corresponds to the cantilever experiencing **net** attractive forces (it can still experience some repulsive forces, but the average force is attractive) and hence is called the attractive solution (Garcia refers to this as the "low amplitude" solution). The other solution has a phase less than 90 and corresponds to net repulsive forces (Garcia calls this "high amplitude") See Fig. 1. In general, the energy dissipated in these two modes is very different. In attractive mode on "hard surfaces" (e.g., crystal surfaces, glass, silicon), attractive mode almost always corresponds to non-contact imaging. The cantilever really turns around before the tip ever "touches" the surface. In this case it isn't so surprising there is considerably less energy dissipation in attractive mode. Fig 1. shows amplitude, phase, and dissipation measured on a hard surface. There is much less dissipation associated with the attractive solution (see also Fig 4.). Also, the dissipation in the attractive region is usually spread over a bigger area because there isn't the small contact area associated with the repulsive interaction. Figures 2 and 3 show a beautiful example measured in Ricardo Garcia's lab of what this can mean in a real imaging situation on some anti-bodies (reproduced here courtesy of the author).
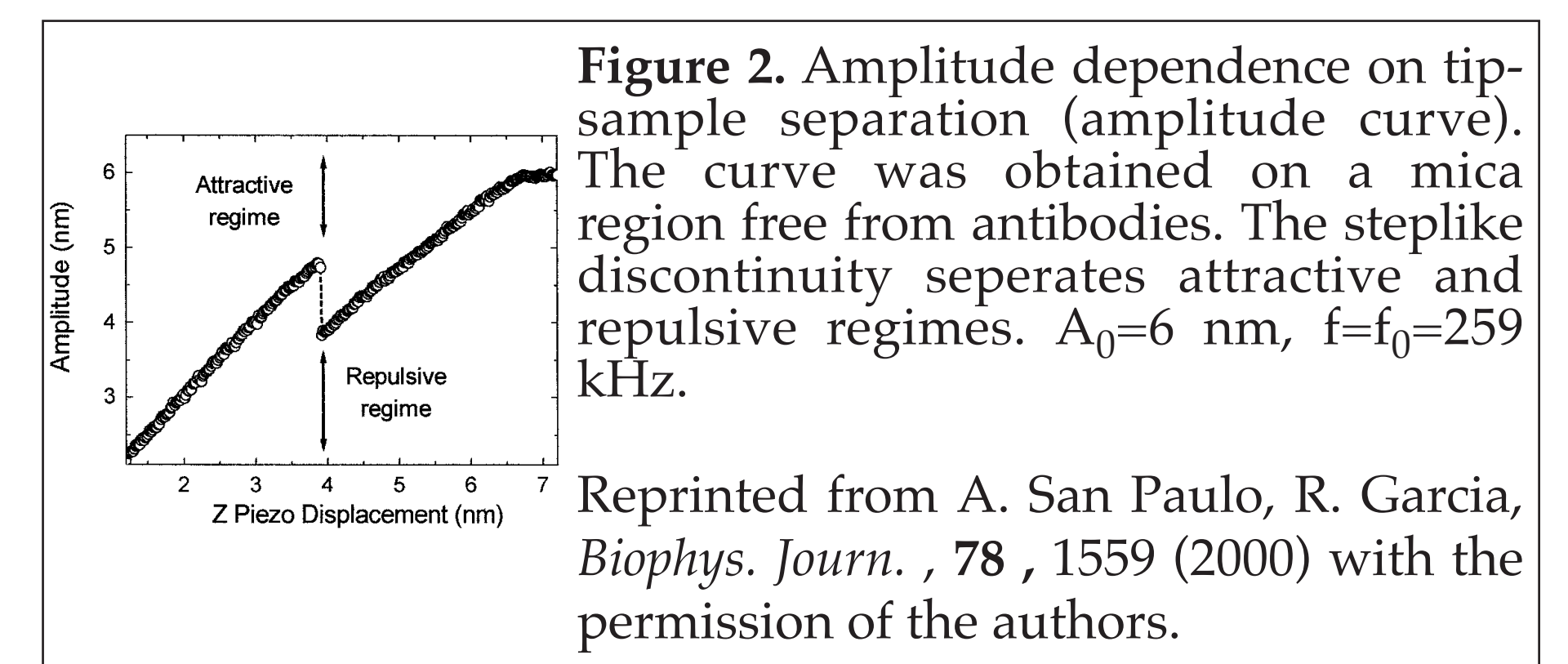

Figure 2. Amplitude dependence on tip-sample separation (amplitude curve). The curve was obtained on a mica region free from antibodies. The steplike discontinuity seperates attractive and repulsive regimes. A_0=6 nm, f=f_0=259 kHz.

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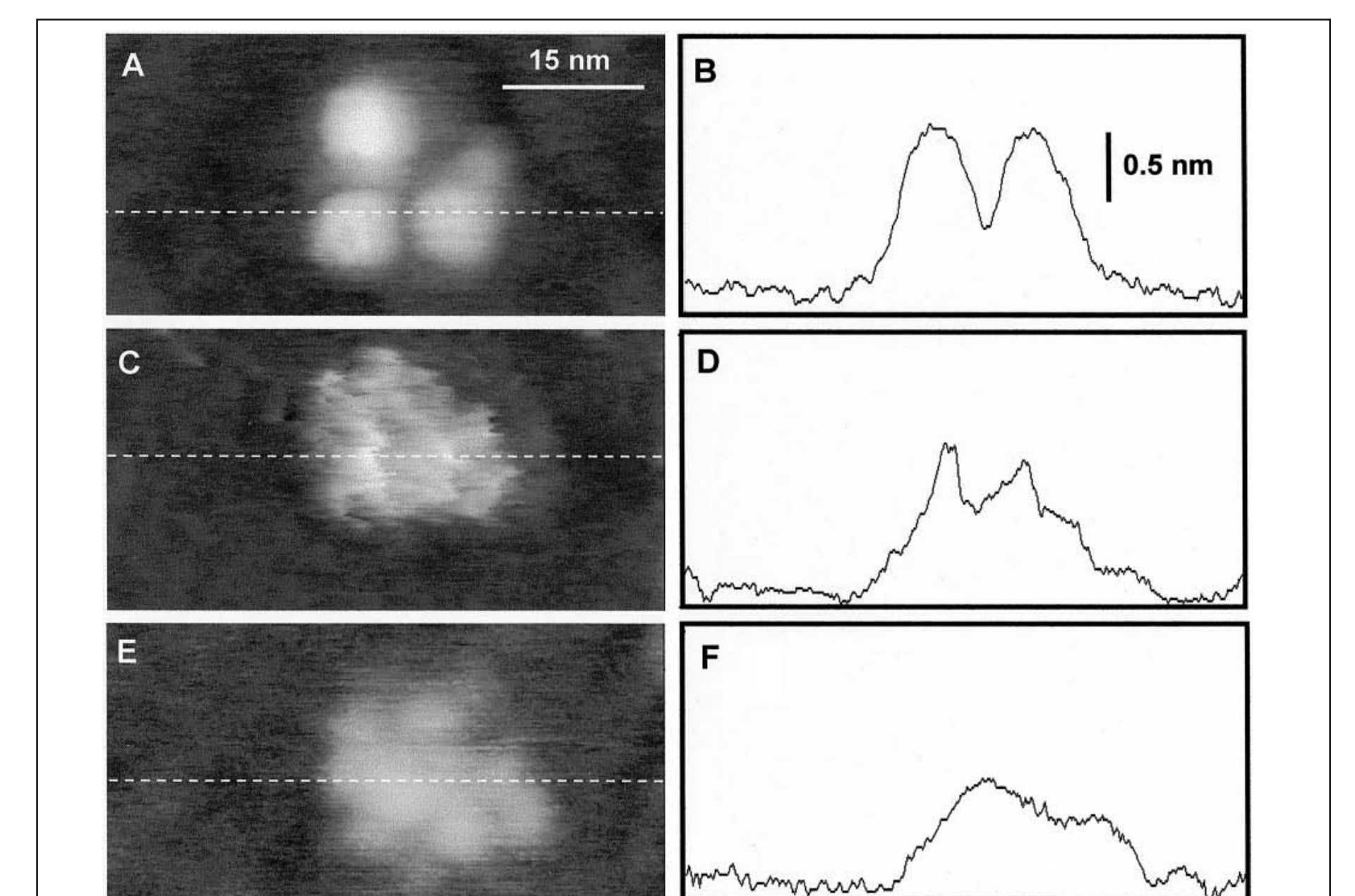

Figure 3. (A) High-resolution tapping-mode AFM image (attractive interaction regime) of a single a-HSA molecule. The three fragments and the hinge regions are clearly resolved. A_{sp}=5.9 nm. (B) Cross-section along the dashed line in (A). (C) The same molecule imaged in the repulsive interaction regime, A_{sp}=2.8 nm. (D) Cross-section along dashed line in (C). (E) Image of the molecule in the attractive interaction regime after repeated imaging in the repulsive regime, A_{sp}=5.9 nm. The comparison between (B) and (F) cross-sections reveals the changes in the topography of the molecule after it was imaged in the repulsive interaction regime. A_0=6 nm and f=f_0=259 kHz in all cases.

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What experimental "knobs" are available to control which mode we operate in?

FIgure 3 is obviously powerful motivation for us to want to control the operating mode we are in, but what parameters can we change to help us accomplish this? The first thing to do is to always be looking at the phase signal while imaging so that you learn to diagnose whether the microscope is running attractive or repulsive. After that, these simple rules (supported by the experimental data in Figs. 4 and 5) will help:

1. Magnitude of the attractive forces
Poor experimental control (except perhaps EFM and MFM). Tip wear is a huge player. Obviously, bigger attractive forces favor attractive mode.

2. Cantilever Quality Factor (Q)
Poor experimental control (except with Q-control). High quality factors favor attractive mode. Low quality factor favors repulsive mode.

3. Drive Frequency
Good experimental control. Driving above resonance favors attractive mode while driving below resonance favors repulsive mode.

4. Free Cantilever Amplitude
Good experimental control. Bigger amplitudes favor repulsive mode. Smaller amplitudes favor attractive mode. Closely related to this is the setpoint. Generally, lowering the setpoint will increase the probability of running in repulsive mode.

References and Acknowledgements

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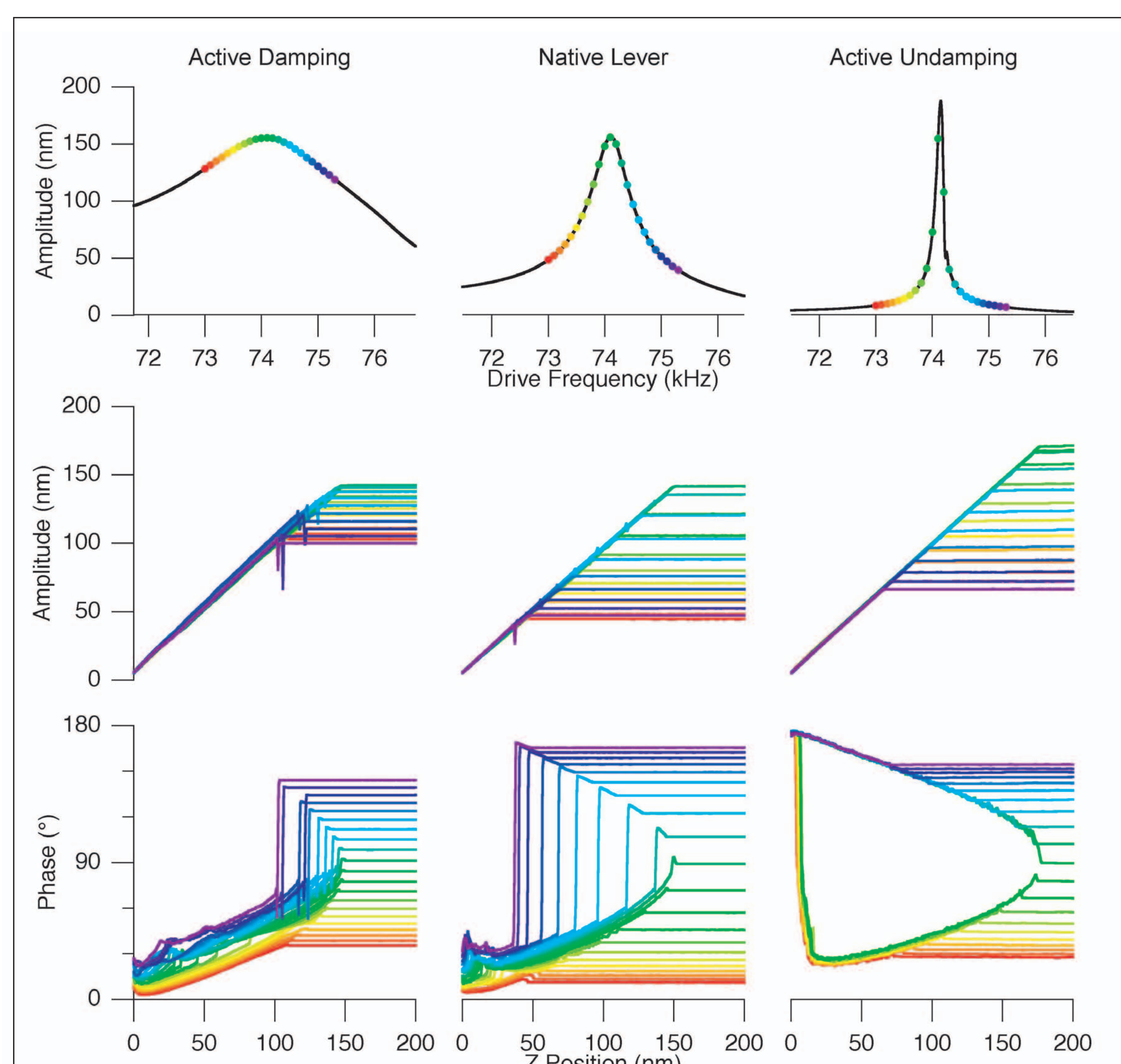
Figure 5. More data than anyone really wants to look at. Amplitude and phase versus distance curves taken at various drive frequencies around the cantilever resonant frequency and at three different quality factors (Q). The Q was changed using digital Q control in the MFP-3D. The data in the left hand column (low Q) is entirely repulsive independent of the drive frequency. The data in the middle column (native Q) is mixed, but driving below the resonance clearly favors the repulsive mode. The right hand column (high Q) is also mixed but has more attractive mode present. Again driving below resonance tends to keep you in repulsive mode. The family of phase curves in the right-hand column nicely shows the two solutions. For small sample and cantilever damping (on hard samples), the theory predicts that the two solutions should lie on an arcsine curve. Indeed, the curves outline a half of a sine wave lying on its side.
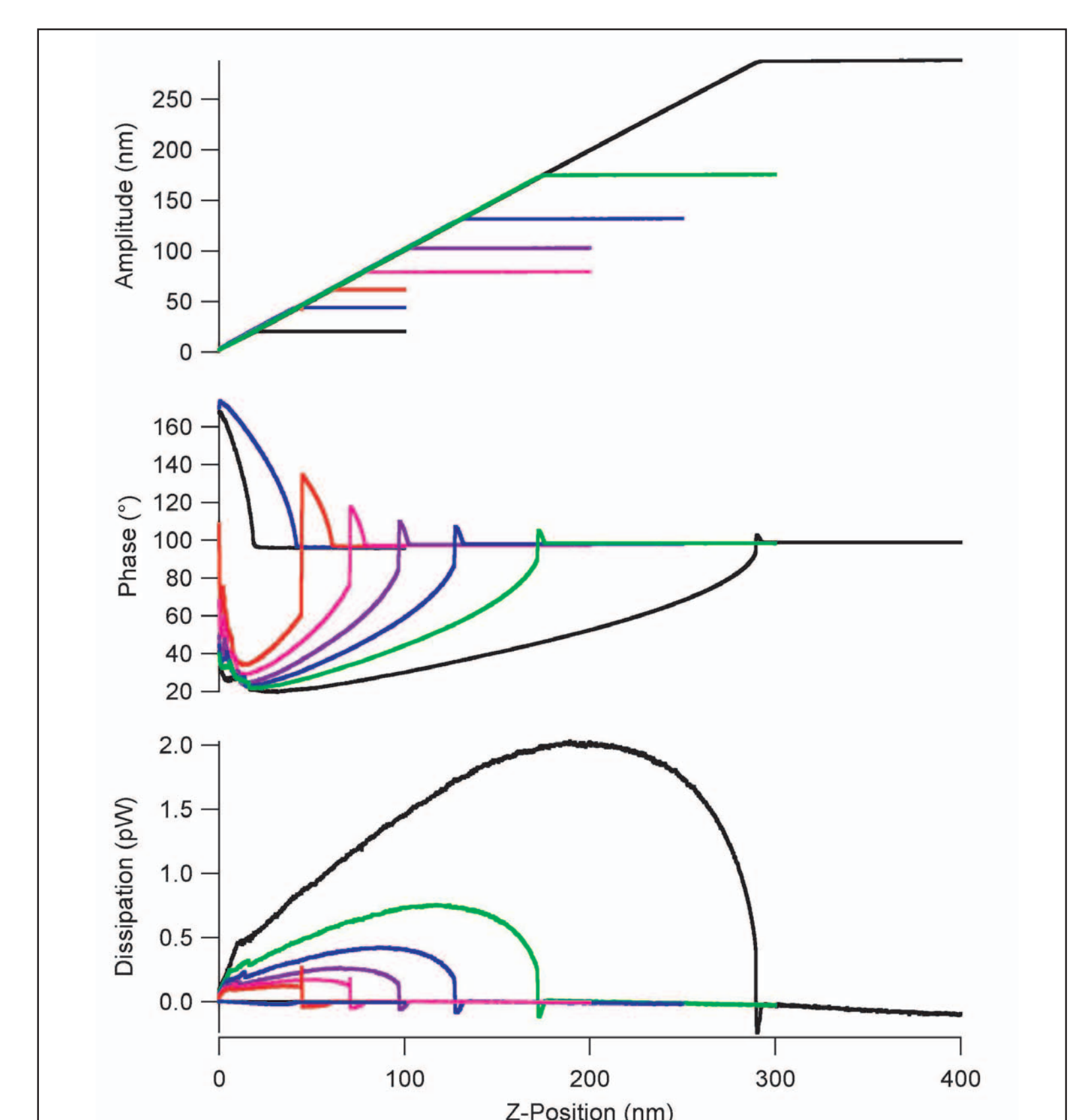

Figure 4. Amplitude, phase, and dissipation vs distance curves illustrating the dependence of the operating mode on the free cantilever amplitude. Starting with larger initial amplitudes curves with almost no attractive regime. Intermediate amplitudes show mixed behavior, while the smallest amplitudes yield purely attractive operation. Data taken with an AC240 Olympus cantilever (k ≈ 3 N/m, f_0 ≈ 70 kHz) on freshly cleaved mica in air.