# Subharmonic Oscillations and Chaos in Dynamic Atomic Force Microscopy

John H. CANTRELL[1], Sean A. CANTRELL[2]
[1]NASA Langley Research Center, Hampton, Virginia 23681, USA
[2]NLS Analytics, LLC, Glencoe, Illinois 60022, USA

**14th International Symposium on Nondestructive Characterization of Materials, June 22-26, 2015, Marina Del Rey, California, USA**

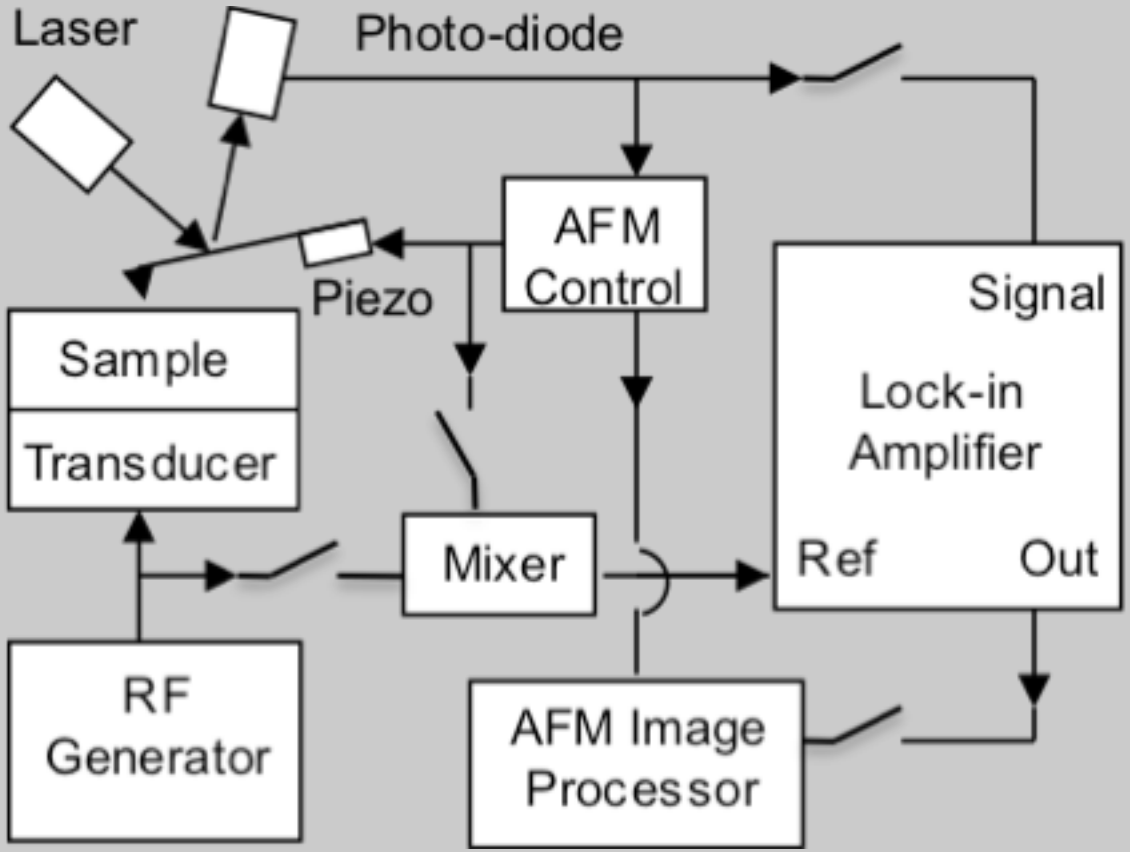
**Explore. Discover. Understand.**

# *Background*

- Atomic force microscopy (AFM) has rapidly evolved from a quasi-static method for assessing material properties at the nanoscale into a fully dynamic technology

- Various dynamic implementations of AFM, collectively called dynamic AFM or *d*-AFM, have been developed to assess surface, near surface, and subsurface properties of the material such as elastic moduli, adhesion, viscoelasticity, embedded particle distributions, device integrity, and topography

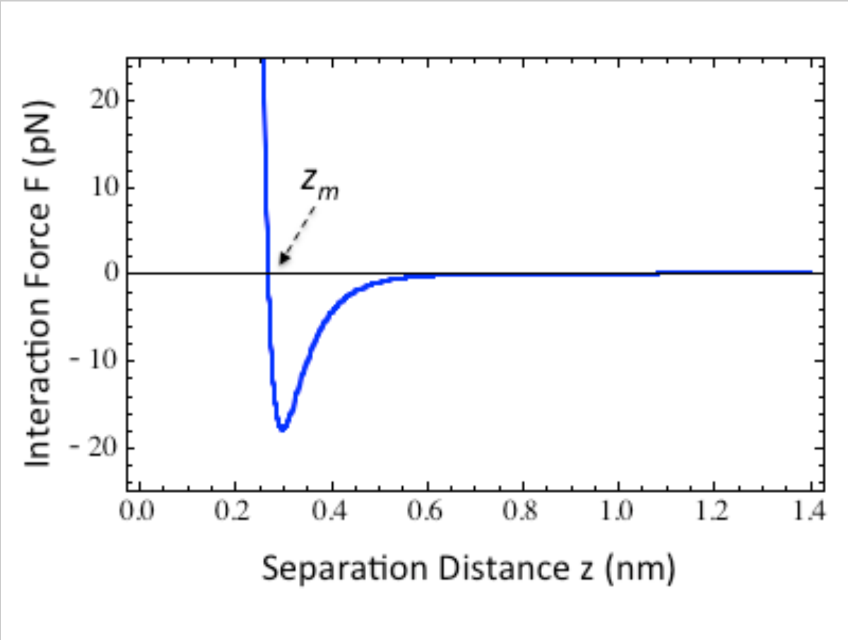
# *Equipment arrangement for various d-AFM techniques using either one or two probing signals*




**Explore. Discover. Understand.**

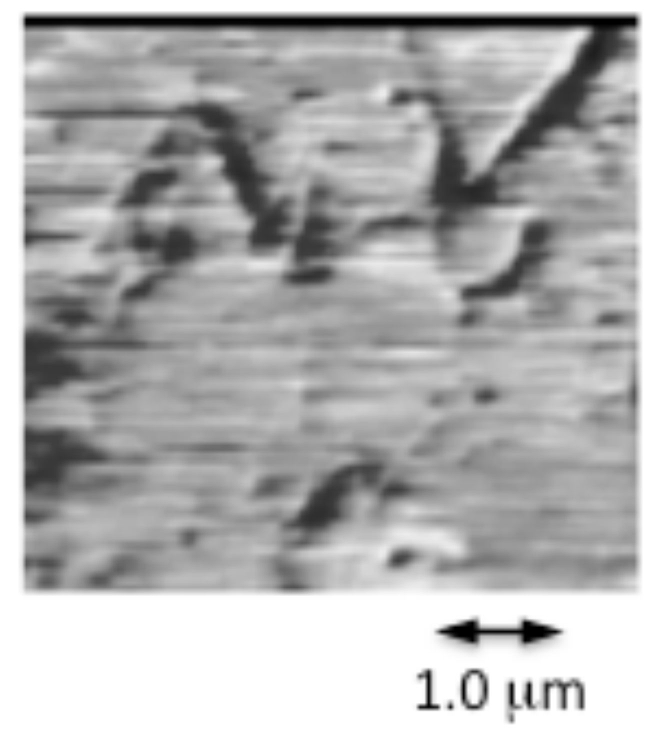
## *Cantilever-sample interaction force*

- The d-AFM output signal is derived from the interaction force between the cantilever tip and the sample surface that is dependent on the tip-sample separation distance z.
- A typical force-separation curve (obtained from Lennard-Jones potential) is shown below



# *d-AFM image degradation*

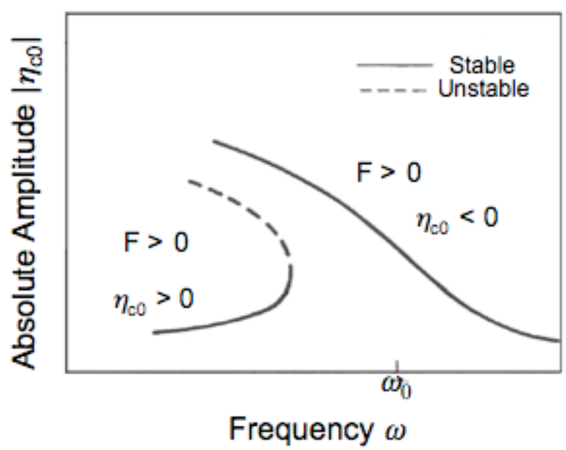
Certain conditions of d-AFM operation generate chaotic oscillations that result in image streaking, as shown below for a RDF-AFUM micrograph of 50mm-thick single wall carbon nanotubes embedded in a LaRC-CP2 polyimide matrix.




**Explore. Discover. Understand.**

# *Bi-stability*

The existence of two stable states of oscillation at certain frequencies (bi-stability) has been proposed as a possible cause of chaotic oscillations, resulting from interaction force nonlinearity.



# *Research objective*

- Develop broad-ranging explanation for chaotic oscillations and image degradation in d-AFM micrographs based on solutions to cantilever dynamical equation that includes quadratic nonlinearity in the interaction force.

- Derive dynamical conditions under which subharmonic generation leading to chaotic oscillations is assured.

# *Cantilever dynamical equations*

The cantilever dynamical motion may be expressed quite generally as an expansion over vibrational modes *n* of the form

$$y(x,t) = \sum_{n=1}^{\infty} Y_n(x)\eta_n(t) \qquad (1)$$

where

$x$ = spatial coordinate $\qquad$ $t$ = time

$Y_n(x)$ = orthogonal basis set of cantilever shape-dependent spatial eigenfunctions

$\eta_n(t)$ = temporal components of the vibrational modes

## *Cantilever dynamical equations*

The temporal components $\eta_n$ of the vibrational modes are obtained from the solutions of

$$m_c \frac{d^2\eta_n}{dt^2} + \gamma_n \frac{d\eta_n}{dt} + k_n \eta_n = F_c \cos\omega t + F\left(z, \frac{dz}{dt}\right) \qquad (2)$$

where

$m_c$ = effective mass of cantilever

$\gamma_n$ = damping coefficient

$k_n$ = cantilever free-space stiffness constant

$F_c$ = cantilever driving force

z = cantilever-sample separation

$F(z, dz/dt)$ = cantilever-sample interaction force

Note: *dz/dt* accounts for the transfer of energy between the cantilever and the sample surface.

# *Interaction force expansion*

Let $z_0$ = the set-point (rest) position of the cantilever tip on the force curve and write $z - z_0 = \eta_n$ for mode n.

Expand the interaction force $F(z,dz/dt)$ as

$$f\left(z, \frac{dz}{dt}\right) = [F_{00} + F_{10}\eta_n + F_{20}\eta_n^2 + \cdots] + \left[F_{01}\frac{d\eta_n}{dt} + F_{02}\frac{d^2\eta_n}{dt^2} + \cdots\right]$$

$$+\left[F_{11}\eta_n\frac{d\eta_n}{dt} + \cdots\right] \qquad (3)$$

$F_{i0}$ ($i$ = 1, 2, 3, ⋯) = 'spring stiffness' coefficients

$F_{0j}$ ($j$ = 1, 2, 3, ⋯) = coefficients that account for the transfer of energy from the cantilever to the sample.

# *Interaction force expansion*

**The interaction force expansion is not a Taylor series.**
Taylor series is limited to small cantilever displacements.

More efficient expansion is obtained by placing interaction force function in the space of smooth functions over entire range of cantilever displacements, equip the space with an inner product defined for arbitrary functions *g* and *h* as

$$\langle g,h \rangle = \int_{-\eta_{\max}}^{\eta_{\max}} \int_{-\dot{\eta}_{\max}}^{\dot{\eta}_{\max}} g(\eta,\dot{\eta})h(\eta,\dot{\eta})\,d\eta\,d\dot{\eta} \qquad (4)$$

where $\eta_{\mathrm{max}}$ and $\eta_{\mathrm{max}}$ dot are the maximum cantilever displacement and the maximum rate of change of displacement, respectively.

# *Expansion coefficients*

To obtain $F_{ij}$, we form the inner product $<(F-f),(F-f)>$ where $F(\eta, d\eta/dt)$ is the actual function and $f$ is the polynomial expansion of $F$ truncated to second order, take derivatives with respect to each of the coefficients $F_{ij}$ in the expansion, set each derivative to zero, and solve the resulting system of linear equations.

For present purposes the most important coefficient is $F_{20}$ given as

$$F_{20} = \frac{15}{16\eta_{\max}^{5}\dot{\eta}_{\max}}\left[3\langle F,\eta^{2}\rangle - \eta_{\max}^{2}\langle F,1\rangle\right] \qquad (5)$$

Previously shown that $F_{20}$ leads directly to bi-stability.

# *Dynamical equation*

Substitute polynomial expansion to second order in the temporal equation for vibrational mode *n*, consider only fundamental mode n = 1, drop the subscript, and divide by $m_c$ to obtain the dynamical equation

$$\frac{d^2\eta}{dt^2} + \Gamma \frac{d\eta}{dt} + \omega_1^2 \eta - \beta \eta^2 = F_m \cos \omega t \qquad (6)$$

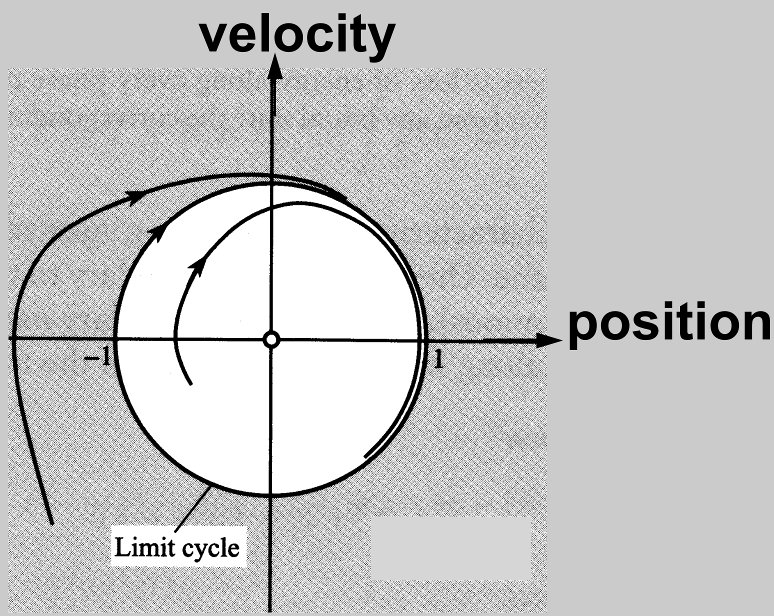
where

$$\Gamma = \left(\gamma - F_{01}\right) / m_c \qquad \omega_1^2 = \left(k - F_{10}\right) / m_c \qquad F_m = F_c / m_c$$

$$\beta = F_{20} / m_c$$

# *The Melnikov method*

- Under certain conditions, solutions to the dynamical equation include subharmonics and chaotic oscillations.
- The Melnikov method provides means to determine threshold conditions necessary to assure a cascade of period doubling bifurcations (subharmonics) leading to chaos in a perturbed nonlinear system by assessing the distance between stable and unstable paths in phase space (velocity-position plots) emanating from an unstable limit cycle (if the limit cycle exists).




**Explore. Discover. Understand.**

# *Threshold conditions for chaos*

- Detailed application of the Melnikov method to the dynamical equation to be published elsewhere.
- Most important result is a quantitative assessment of the threshold drive amplitude that assures a cascade of frequency-halving bifurcations (i.e., subharmonics $\omega/2$, $\omega/4$, $\omega/8$, $\cdots$ of the cantilever drive frequency $\omega$) terminating in chaos.
- The threshold cantilever drive force per unit mass $(F_m)_{th}$ given as

$$\left(F_m\right)_{th} = \frac{1}{5\pi}\frac{\Gamma}{\beta}\left(\frac{\omega_1^5}{\omega^2}\right)\sinh\left(\frac{\omega\pi}{\omega_1}\right) \tag{7}$$

Note dependence on parameter of nonlinearity $\beta$.

# $\eta_{max}$ , $(d\eta/dt)_{max}$ dependence of $(F_m)_{th}$

- Threshold condition $\left(F_m\right)_{th} = (\Gamma\omega_1^5 / 5\pi\omega^2\beta)\sinh(\omega\pi / \omega_1)$ assures the generation of a cascade of period doubling bifurcations leading to chaos as drive amplitude $F_m$ monotonically increased above $(F_m)_{th}$.
- $(F_m)_{th}$ depends on the cantilever drive frequency $\omega$, the natural oscillation frequency $\omega_1$, the damping/energy transfer coefficient $\Gamma$, and the nonlinearity parameter $\beta$ of the interaction force.
- Recall $\beta = F_{20}/m_c$ and $F_{20} = \left(15 / 16\eta_{\max}^5\dot{\eta}_{\max}\right)\left[3\langle F,\eta^2\rangle - \eta_{\max}^2\langle F,1\rangle\right]$ .

,

- Conclude $(F_m)_{th}$ depends on maximum displacement of the cantilever from its set-point and the maximum rate of change of the displacement over oscillatory cycle.

# *Conditions for scan instability*

- Quantitative assessment of threshold drive force $(F_c)_{th} = m_c(F_m)_{th}$ can be calculated from the Lennard-Jones potential

$$\phi_{LJ} = \phi_0 \left[ \left( z_m / z \right)^{12} - \left[ \left( z_m / z \right)^{6} \right] \right] \qquad (8)$$

where $\phi_0 = 4.05 \times 10^{-22}$ J and $z_m = 0.27$ nm.

- The interaction force $F(z, dz/dt) = F(\eta, d\eta/dt) = -(d\phi_{LJ}/dz)$.

- Substitute $F(\eta, d\eta/dt)$ in the expression for $F_{20}$

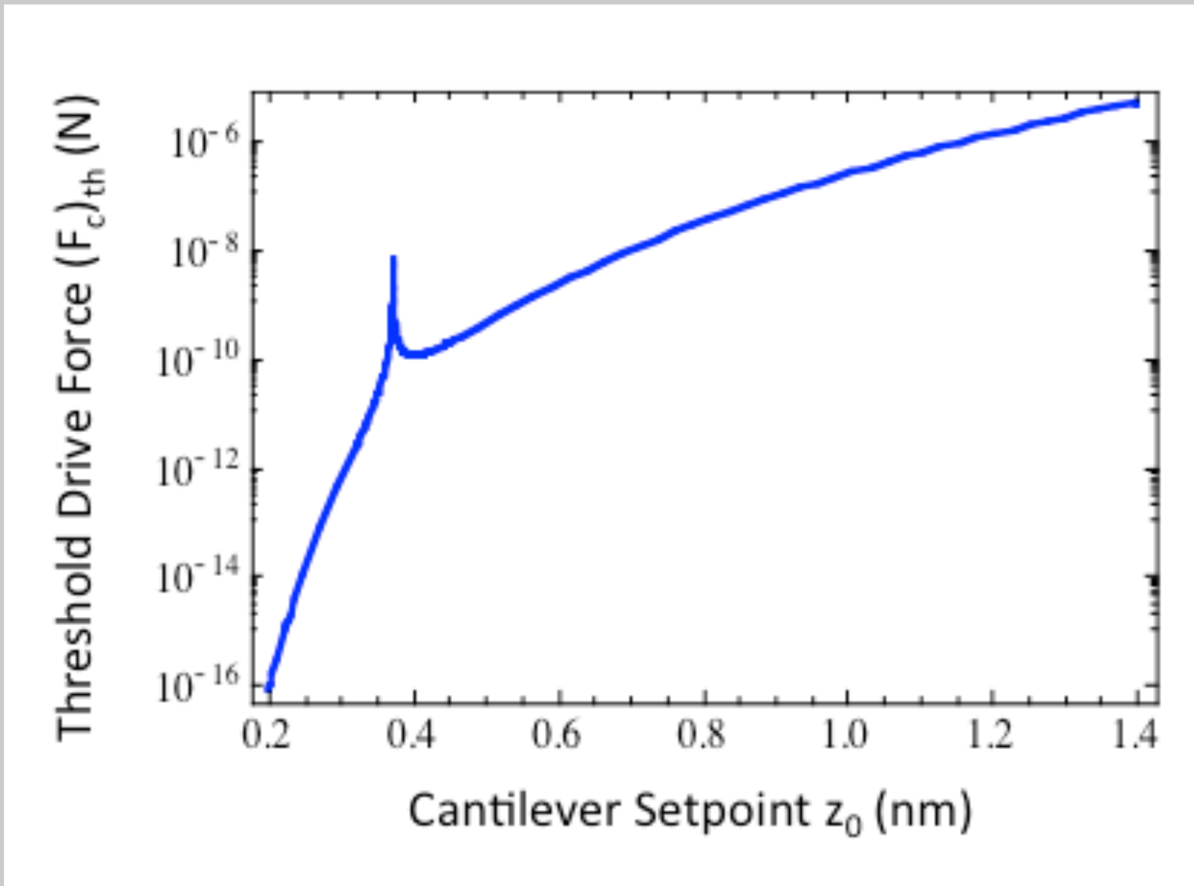
$$F_{20} = \frac{15}{16\eta_{\max}^5 \dot{\eta}_{\max}} \left[ 3\langle F, \eta^2 \rangle - \eta_{\max}^2 \langle F, 1 \rangle \right] \qquad (9)$$

to obtain $\beta = F_{20}/m_c$

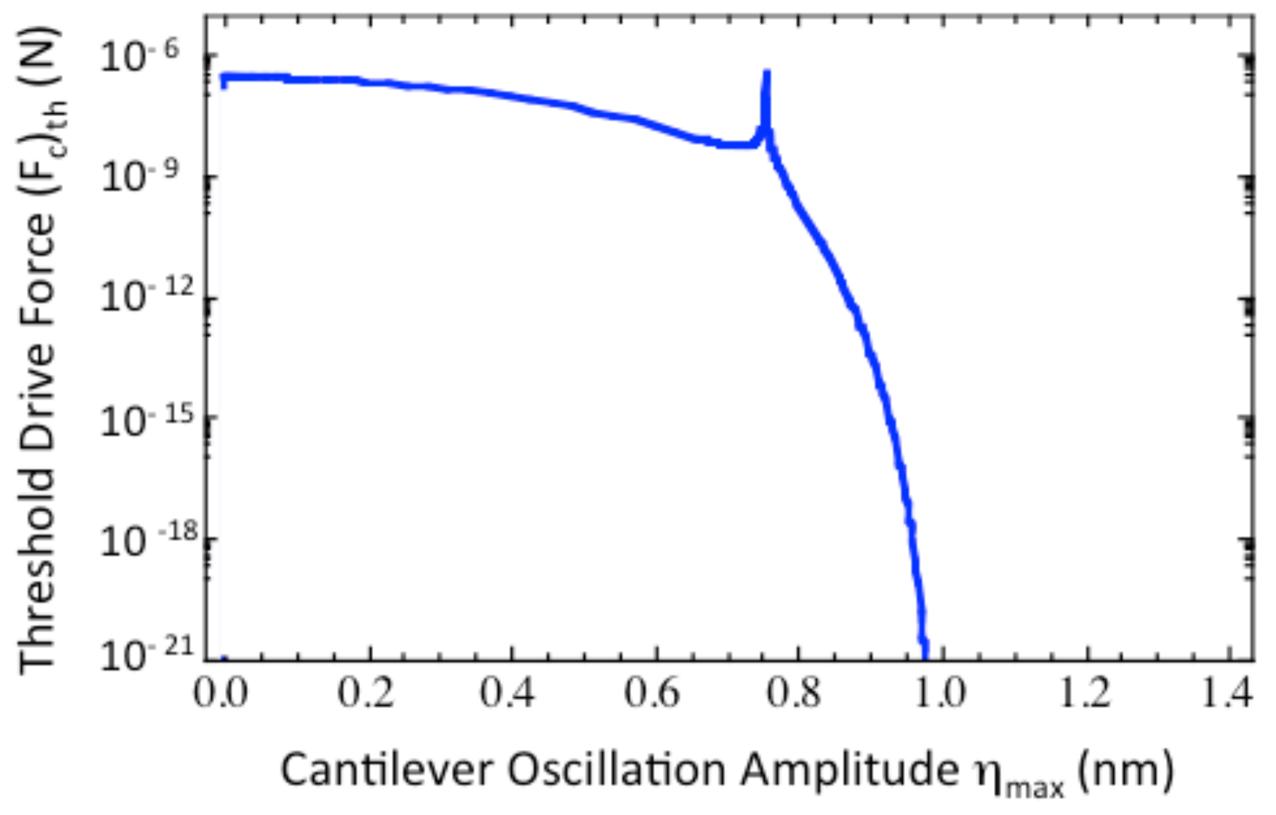
- Calculate $(F_c)_{th}$ from threshold equation

$$\left(F_c\right)_{th} = m_c \left(F_m\right)_{th} = m_c (\Gamma \omega_1^5 / 5\pi\omega^2 \beta) \sinh(\omega\pi / \omega_1) \qquad (10)$$

## *Log-linear graph of $|(F_c)_{th}|$ versus setpoint $z_0$ for oscillation displacement amplitude $\eta_{max}$ = 0.1nm.*

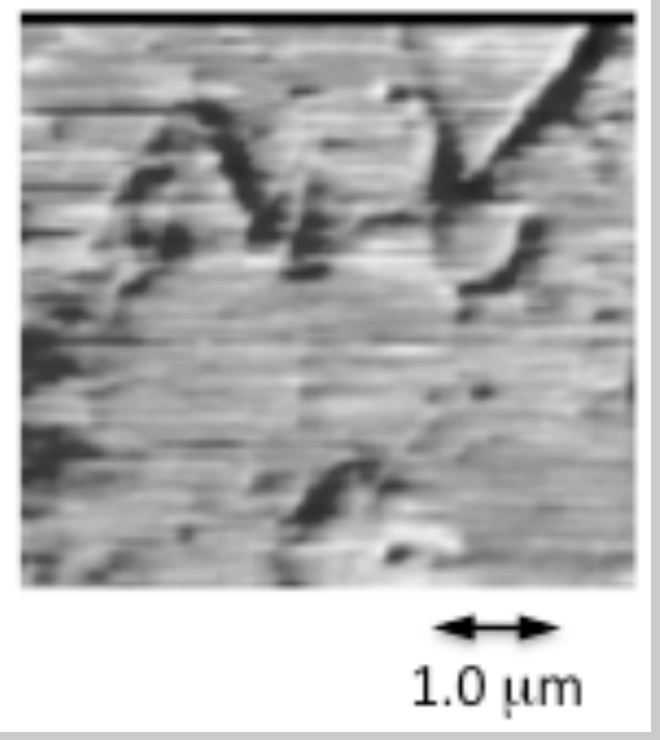


# *Log-linear graph of $(F_c)_{th}$ versus oscillation displacement amplitude $\eta_{max}$ for fixed set-point $z_0 = 1.0$ nm*



# *RDF-AFUM image degradation*

**RDF-AFUM micrograph of 50mm-thick single wall carbon nanotubes embedded in a LaRC-CP2 polyimide matrix.**



# *Chaos in d-AFM*

- Present model applies to all commonly used *d*-AFM modalities.
- For example, contact models of cantilever dynamics show that chaotic oscillations occur when the cantilever makes contact with a hard sample surface - a problem of special importance for AM-AFM (tapping mode).
- Present model based on shape of the force curve as a continuous curve at the atomic scale,
- 'Contact' in model occurs when the cantilever set-point is in the more linear region of the interaction force curve but the amplitude of cantilever oscillation is sufficiently large to produce an excursion into the highly nonlinear region in the vicinity of the ample surface and the minimum of the curve.
- The large amplitude excursion produces a dramatic increase in the value of $\beta$ and a decrease in $(F_m)_{th}$ that results in chaotic oscillations in AM-AFM.

# *Conclusion*

- Developed explanation for chaotic oscillations and image degradation in *d*-AFM micrographs based on solutions to cantilever dynamical equation that includes quadratic nonlinearity in the cantilever-sample interaction force.

- Represented interaction force as polynomial expansion with coefficients that account for the interaction stiffness parameter, the cantilever-to-sample energy transfer, and the amplitude of cantilever oscillation.

- Derived dynamical conditions under which period doubling bifurcations leading to chaos in *d*-AFM is assured.