# High-Frequency PFM and Advanced Imaging Modes

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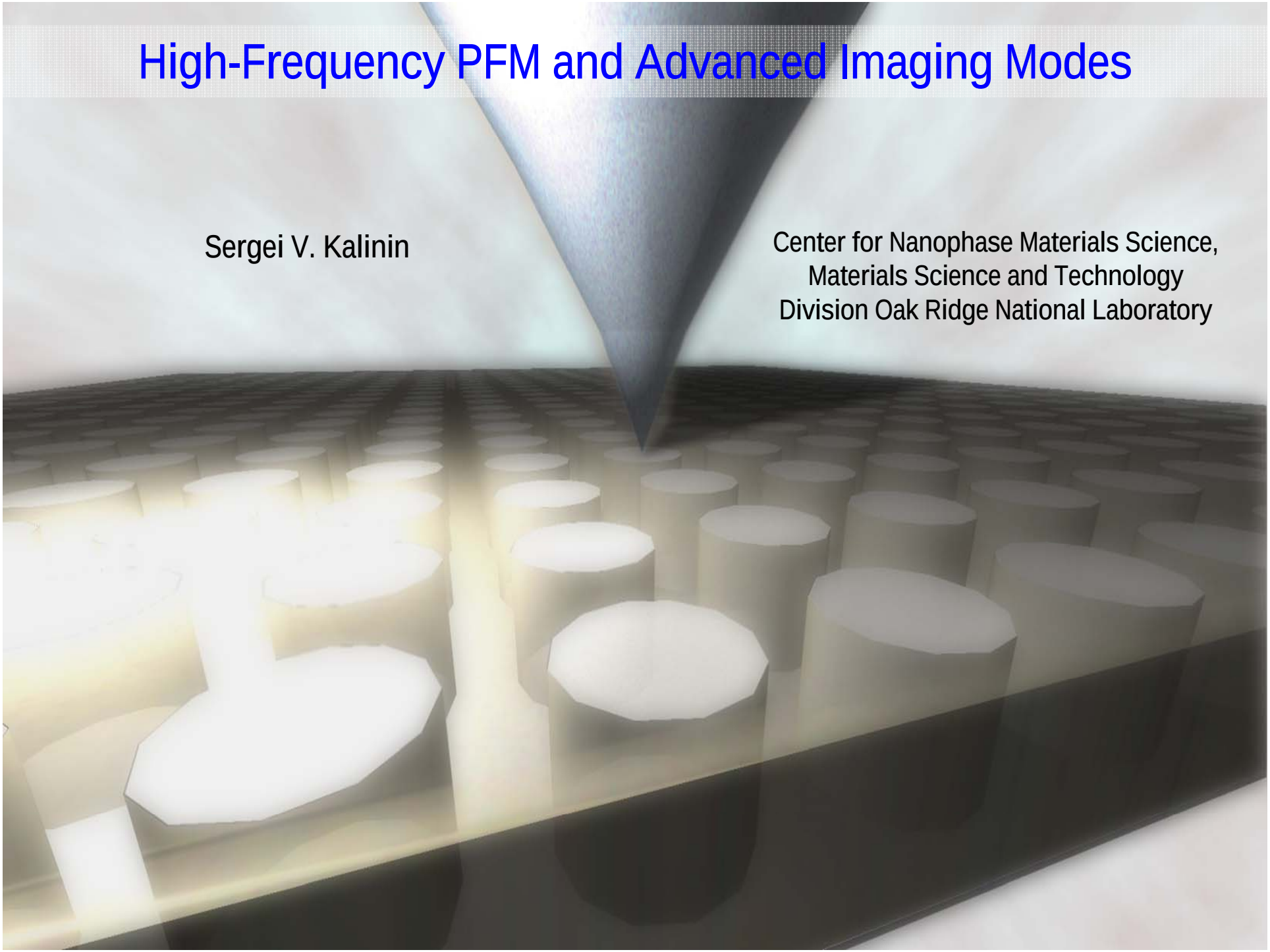
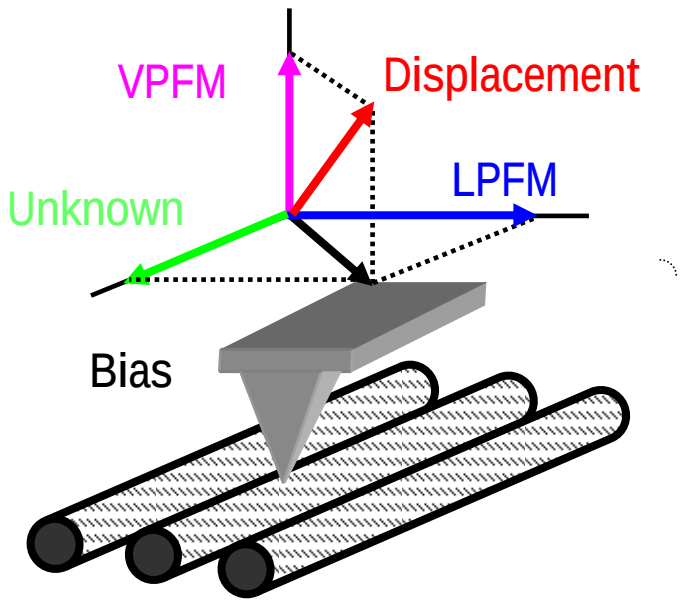


# Dynamics in PFM




Several competing contributions:

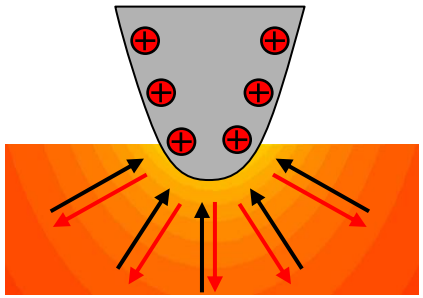
1. Vertical surface response
2. Longitudinal response
3. Torsional response
4. Local electrostatic force
5. Distributed electrostatic force

General calibration:

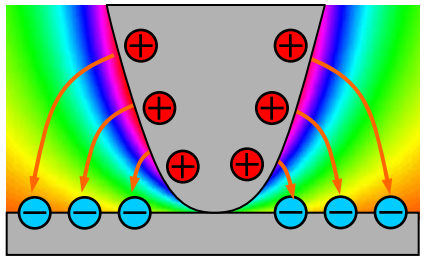
$$\begin{pmatrix} xPR_v \\ xPR_l \\ yPR_v \\ yPR_l \end{pmatrix} = \begin{pmatrix} c & 0 & a \\ 0 & b & 0 \\ 0 & c & a \\ b & 0 & 0 \end{pmatrix} \begin{pmatrix} w_1 \\ w_2 \\ w_3 \end{pmatrix} V_{ac} + \begin{pmatrix} 0 \\ q \\ 0 \\ q \end{pmatrix} V_{ac}\left(V_{dc} - V_{surf}\right)$$

Different contributions to signal can be distinguished by frequency-dependent measurements; however, this approach is not universal

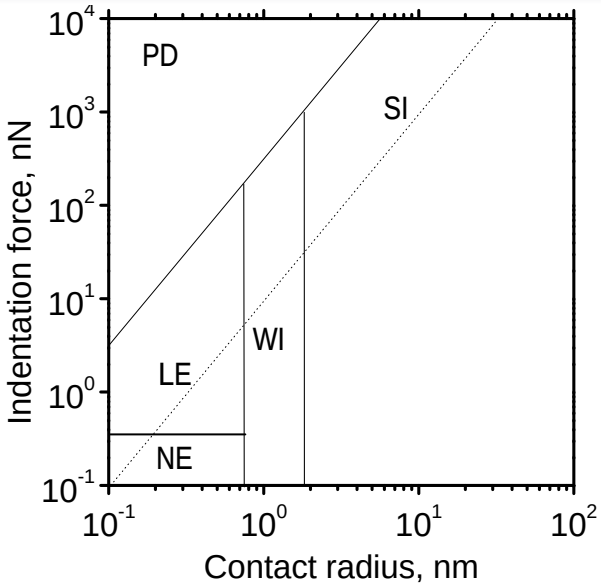
# Contributions to Vertical PFM Contrast



Local Piezoeffect



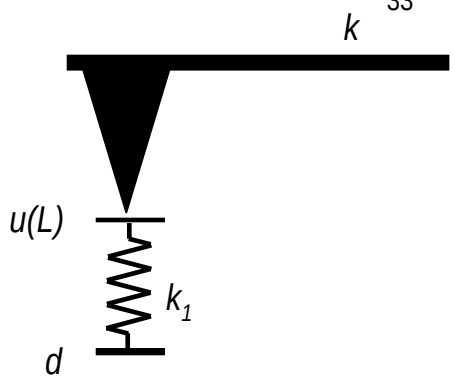
Local Electrostatic Force



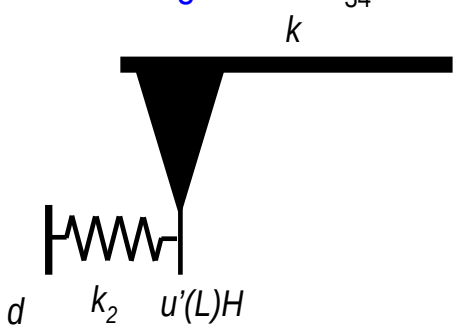

Kalinin and Bonnell, PRB 2001

- Electromechanical response of the surface and local electrostatic force operate in tip-surface junction.
- In a realistic system, longitudinal tip displacement and distributed electrostatic forces contribute to vertical PFM signal

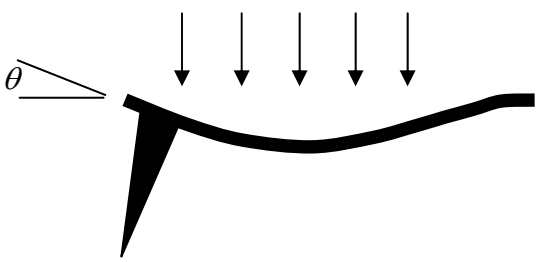
Vertical: Electrostatics + $d_{33}$




Longitudinal: $d_{34}$

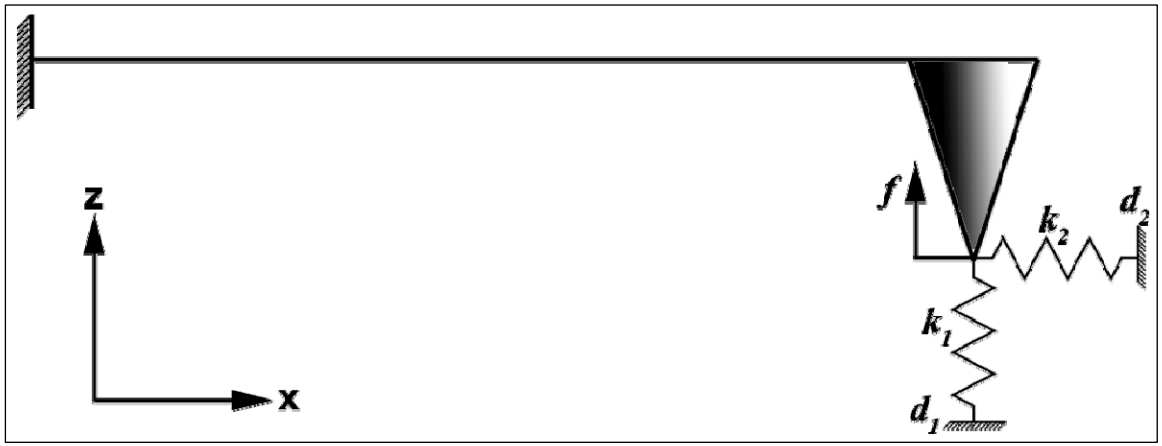



Non-local electrostatic




Vertical PFM signal is a sum of 4 contributions dependent on frequency and geometric properties of the cantilever, and contact stiffness.

# General Solution for Beam Equation




Beam equation:

$$\frac{d^4u}{dx^4} + \frac{\rho S_c}{EI}\frac{d^2u}{dt^2} = \frac{q(x,t)}{EI}$$

Boundary conditions:

$$EIu_0''(L) = k_2 H\left(\tilde{d}_2 - u_0'(L)H\right)$$

$$EIu_0'''(L) = -f_0 + k_1\left(u_0(L) - d_1\right)$$

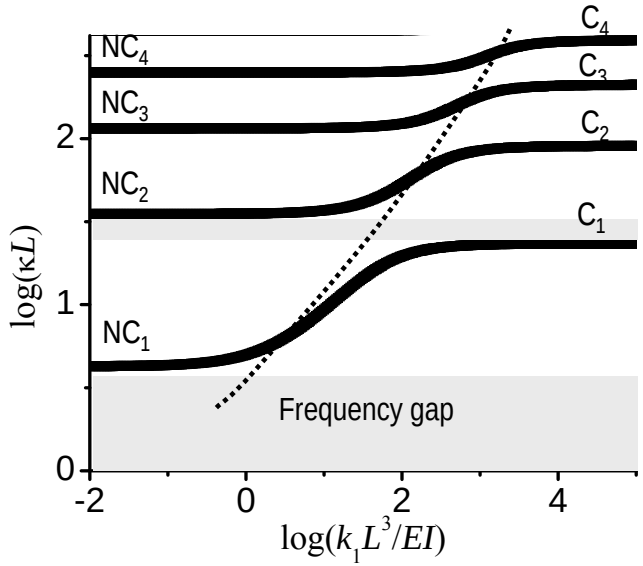
General solution:

$$\theta_{tot} = \frac{A_v(\beta)d_1 + A_l(\beta)d_2 + A_e(\beta)f_0 + A_q(\beta)q_0}{N(\beta)}$$

- Superposition of vertical, longitudinal, local electrostatic, and non-local electrostatic terms
  $\Rightarrow$ can be analyzed separately
- Resonances are determined by the elastic properties of material only
  $\Rightarrow$ can not be used to separate responses
  $\Rightarrow$ AFAM measurements with electric excitation
- Antiresonances can be used to nullify chosen response component

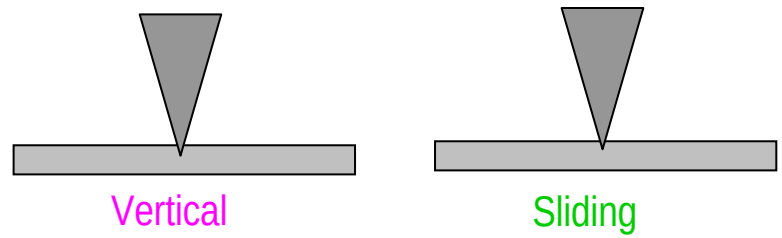
# General Solution for Beam Equation

## Resonance structure




- Well studied in the context of AFAM
- Resonances depend only on elastic properties of material
- But: for non-planar tip spring constant (and resonant frequency) are bias dependent!
- The optimal detection regime depends on mode number and ratio of cantilever spring constant and spring constant of tip-surface junction

## Cantilever geometry




$$\frac{A_v(\beta)}{N(\beta)} \approx \frac{6kLd_{33}}{4kL^2 + 3H^2k_2}$$

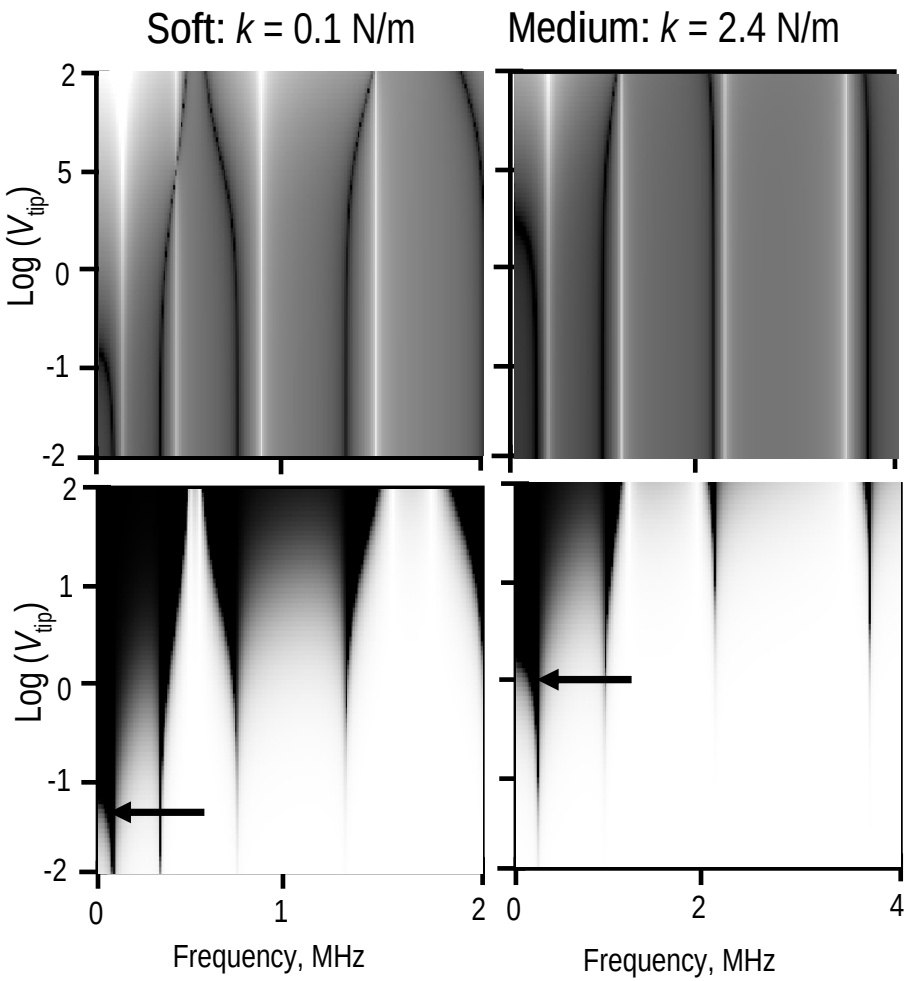
- Depending on the geometric parameters of the cantilever, response can be vertical and lateral

## Frequency behavior

| | |
|---|---|
| Longitudinal: | $\frac{A_l(\beta)}{N(\beta)} \sim \frac{1}{\omega^{1/2}}$ |
| Vertical and local electrostatic | $\frac{A_v(\beta)}{N(\beta)} \sim \frac{A_e(\beta)}{N(\beta)} \sim \frac{k_1}{\omega}$ |
| Non-local | $\frac{A_q(\beta)}{N(\beta)} \sim \frac{1}{\omega^{3/2}}$ |

- All components decrease with frequency
- Non-local decays fastest
- Longitudinal – slowest, but there is an onset of sliding friction

# Cantilever Buckling Effect

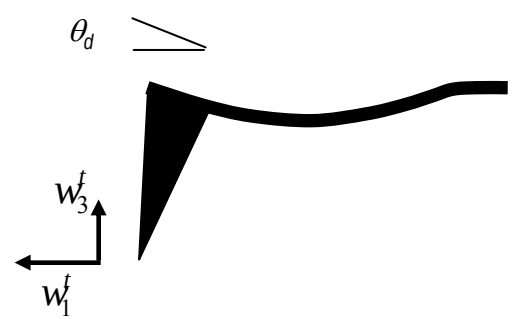



PFM signal: $z_{1\omega} = z_{eff}^{0} + d_{33}V_{ac}$

Effective deflection: $z_{eff}^{0} = -Lw\varepsilon_0 V_{ac}\Delta V \big/ 48k_{eff}H^2$

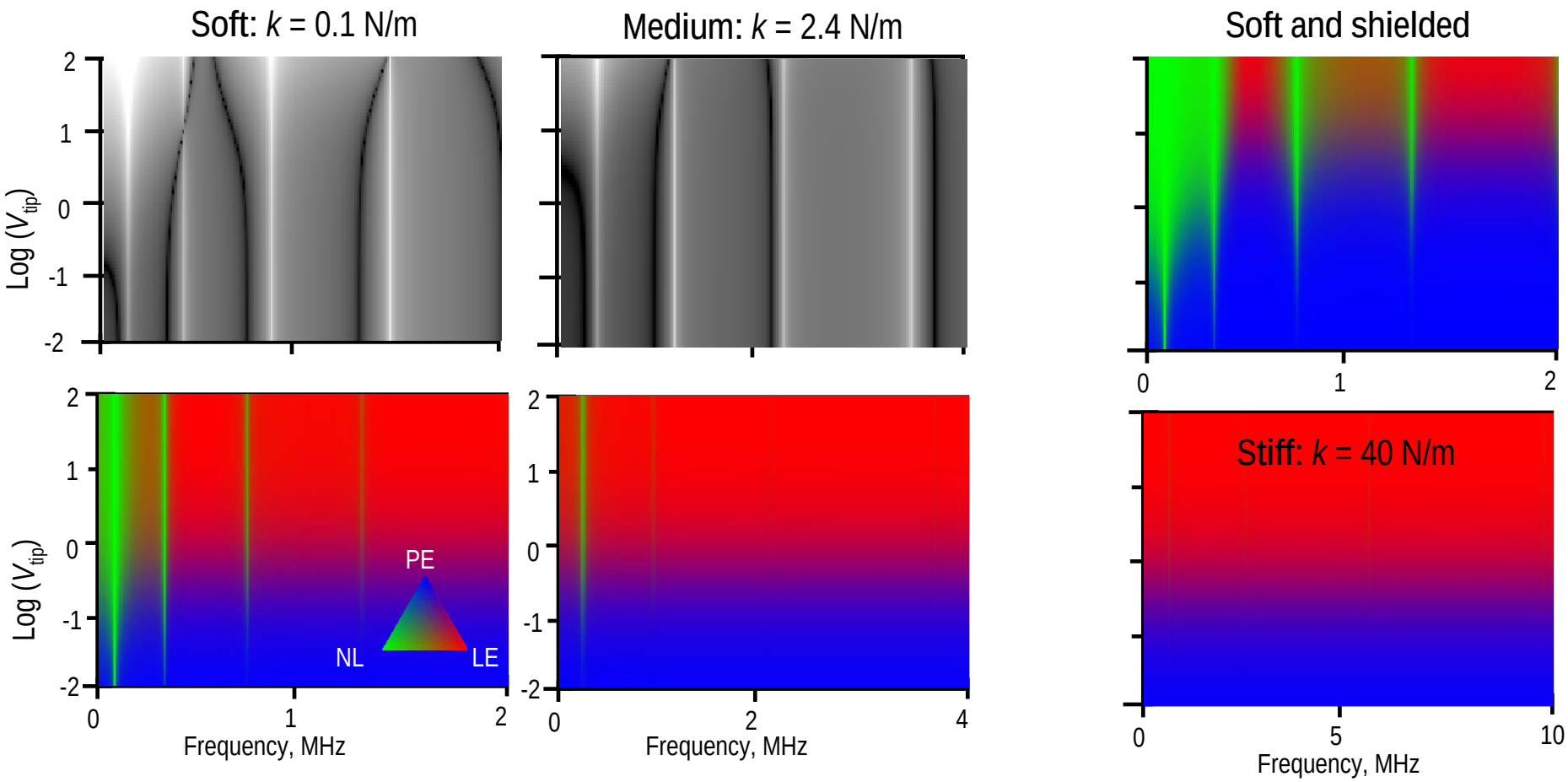
## Which cantilever is stiff enough:

$$d_{33}V_{ac} >> z_{eff}^{0} \quad \Rightarrow \quad k_{eff} >> k^* = \frac{Lw\varepsilon_0\Delta V}{48d_{eff}H^2}$$




- Use of stiff cantilevers
- Positioning the beam and the nodal point for flexural mode (B. Huey)
- Imaging at high frequencies

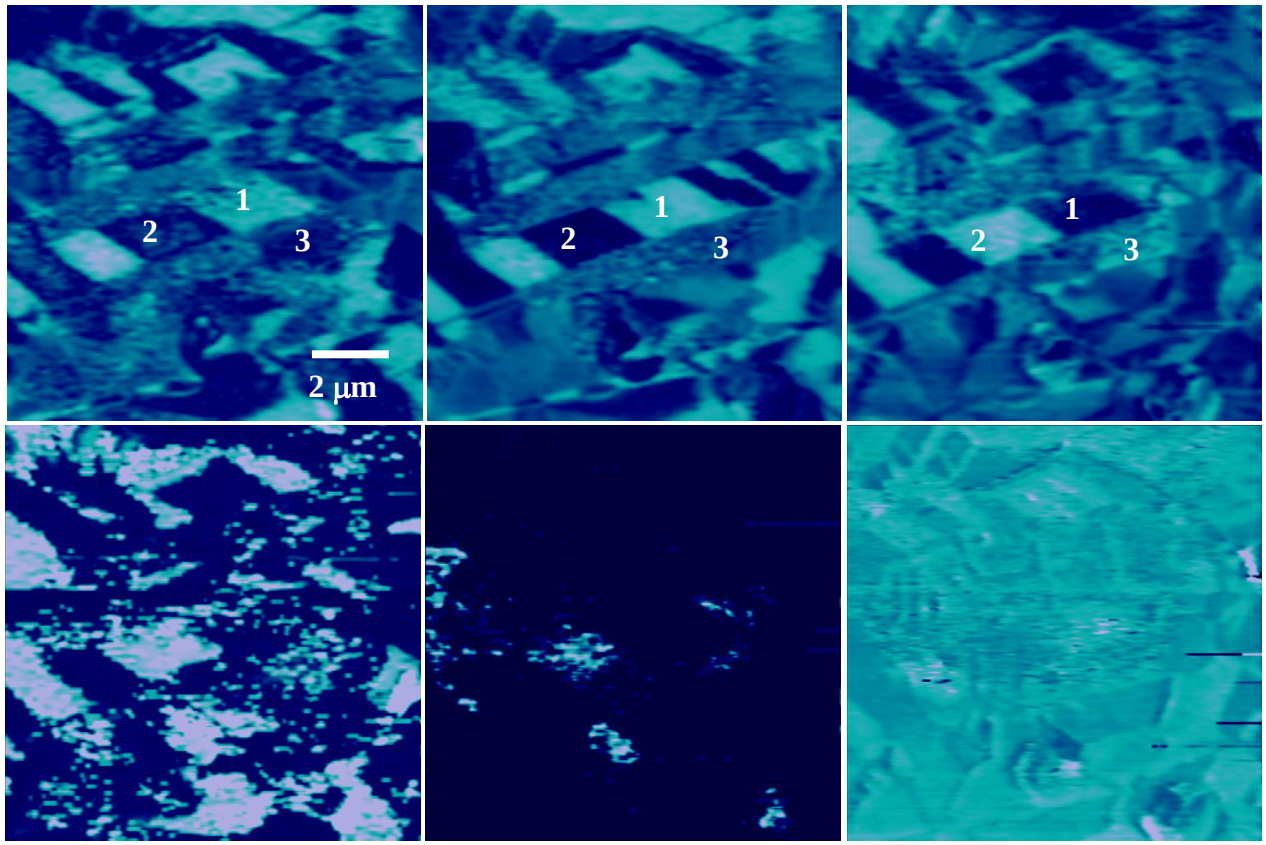
# Cantilever Buckling Effect

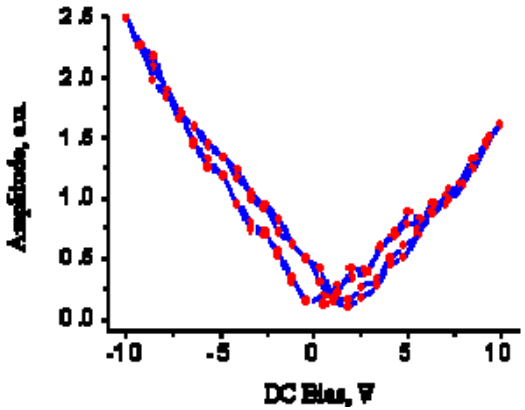


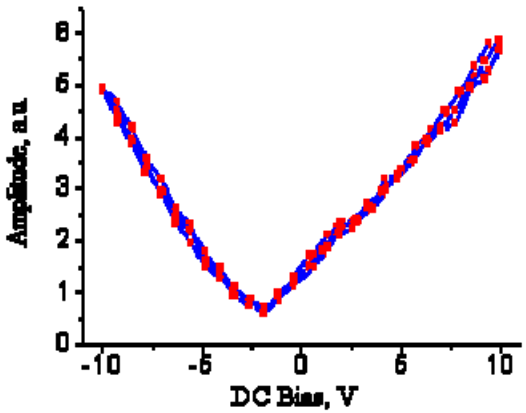

- The relative contributions of the responses do not change near the resonances
- Non-local electrostatics dominate near antiresonances
- Cantilever stiffness and frequency do not affect relative local electrostatic and piezoelectric signal
- Contact stiffness controls resonance frequency

# Electrostatic Effect on Imaging

$V_{tip}$ = -10 V $V_{tip}$ = 0 V $V_{tip}$ = 10 V

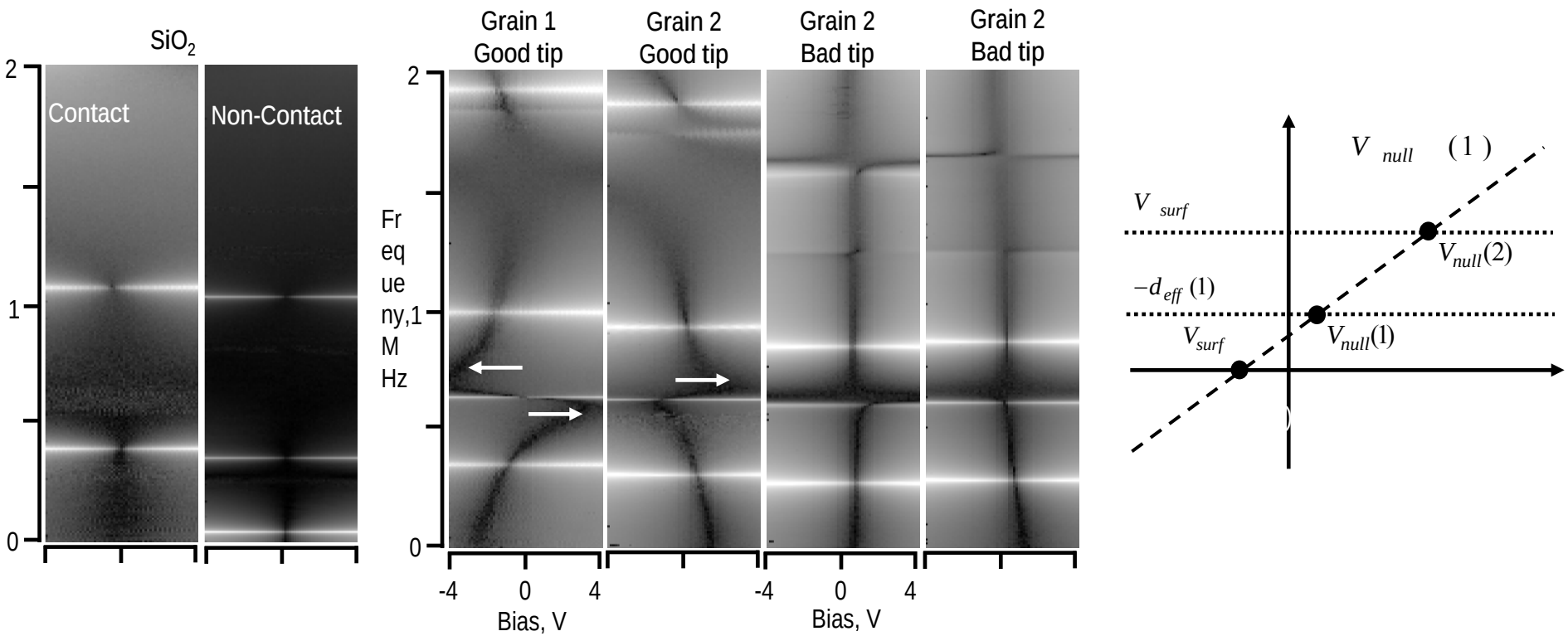


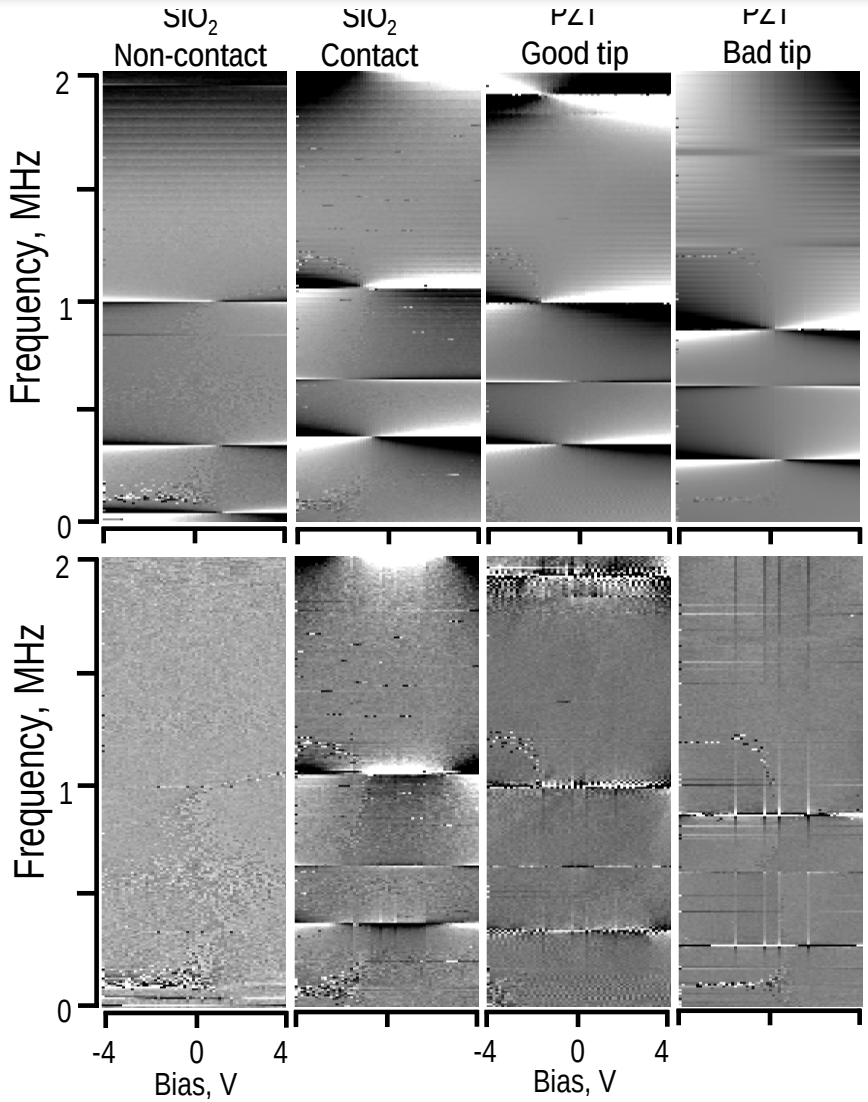

- Electrostatic contribution can strongly affect apparent contrast
- For non-piezoelectrics, yields V-type hysteresis loops
- For switchable ferroelectrics, can be subtracted (since it is conservative)
- For piezoelectrics, difficult to separate

# 2D Spectroscopy

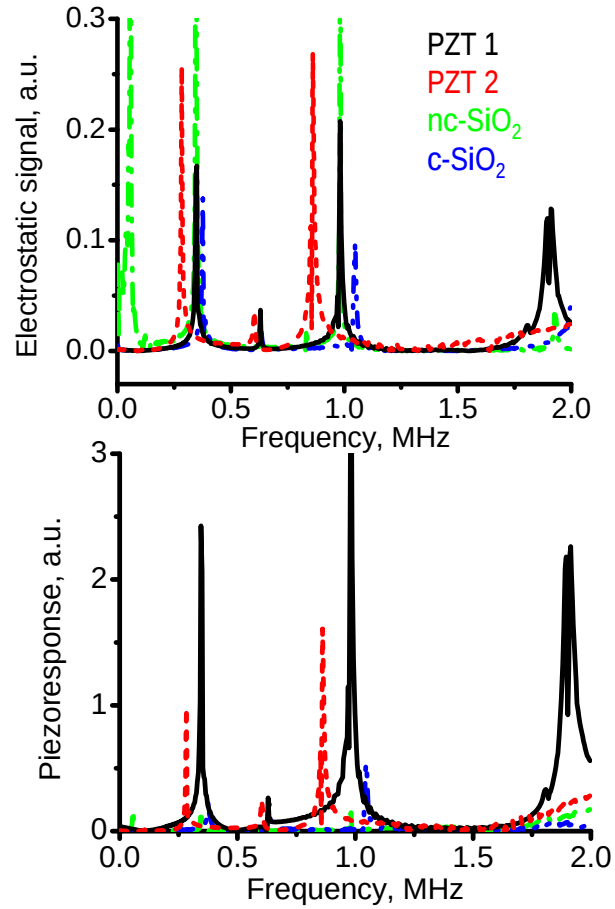



- Frequency dependence of nulling potential is a measure of relative electrostatic and electromechanical contributions to the signal
- This is also valid for NC-AFM based Kelvin probe

# Separation of Electrostatic and Electromechanical Signals

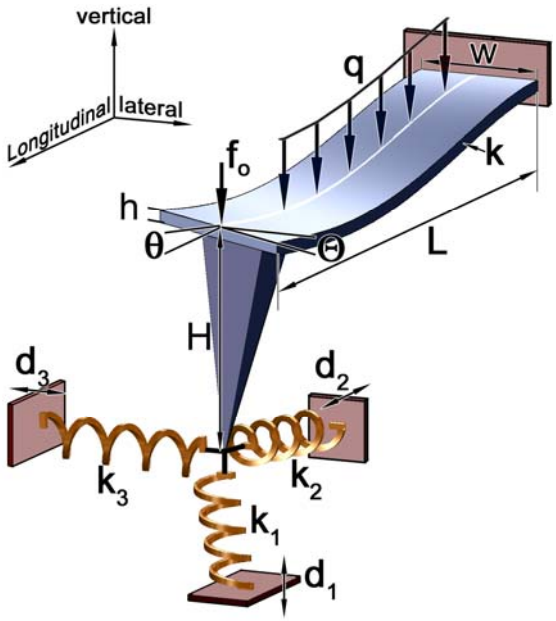



$$PR_{+} = \tilde{d}_{eff} + \tilde{G}_{elec}\left(V_{dc} - V_{surf}\right)$$




- Electrostatic contribution can be unambiguously determined as a function of frequency
- Electromechanical contribution can be determined only if surface potential is known

# Low Frequency PFM Equation




PFM Signal in the Low-Frequency Limit:

$$PR = \alpha_a(h)\tilde{d}_{33}\frac{k_1}{k_1+k} + \frac{C'_{sphere}+C'_{cone}}{k_1+k}(V_{dc}-V_s) + \frac{C'_{cant}}{24k}(V_{dc}-V_{av})$$

Non-local electrostatic contribution

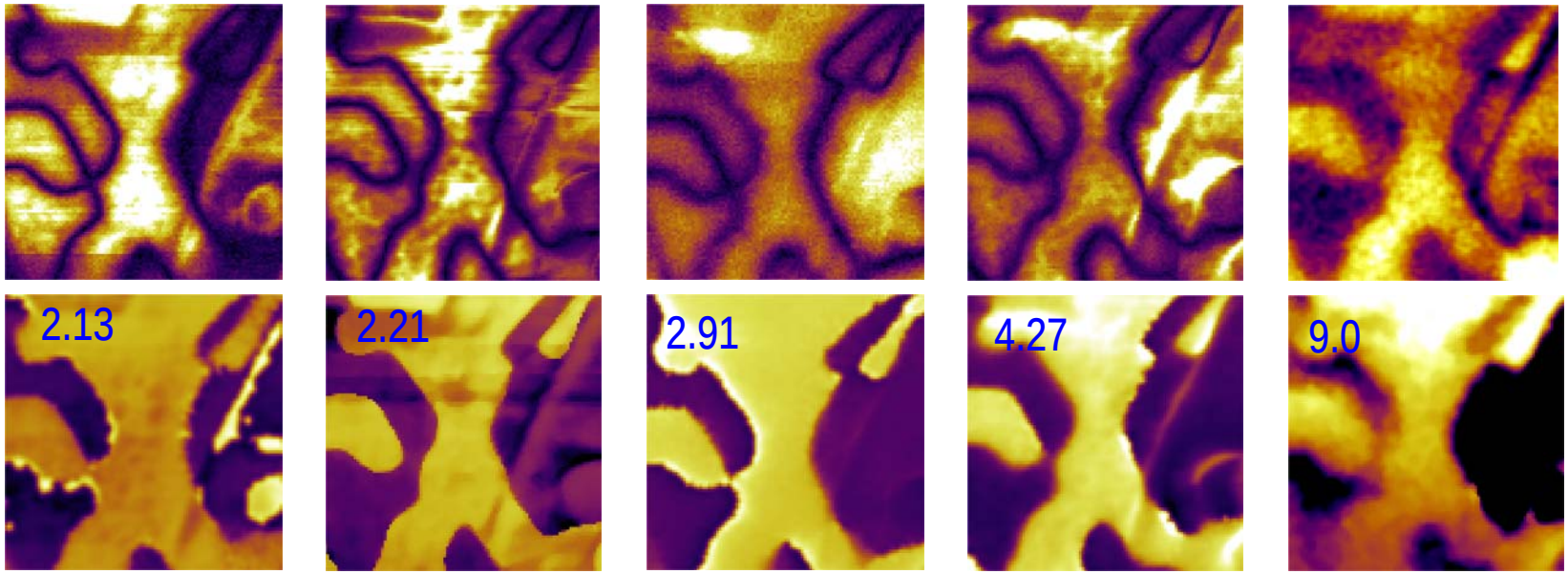
Local electrostatic contribution

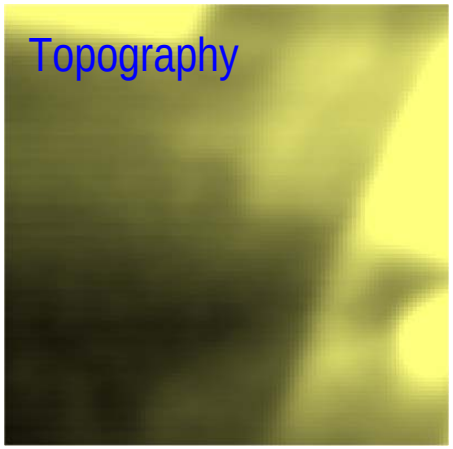
Electromechanical contribution

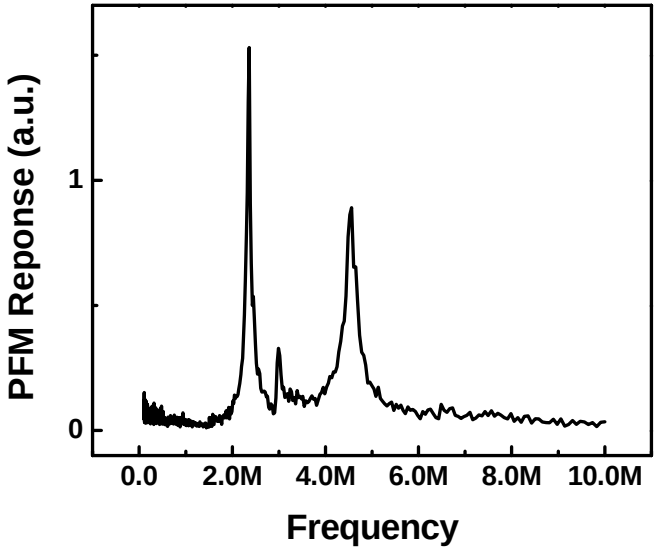
Electromechanical imaging requires high contact stiffness

- Vertical PFM is optimal at high (~1 MHz) frequencies
  - better S/N ratio
  - cantilever stiffening improves contact
  - limited by bandwidth of detector
  - at very high frequencies, no signal transduction
- Lateral PFM is optimal at 10 – 100 kHz
  - problems with friction

# Optimal Frequency for PFM



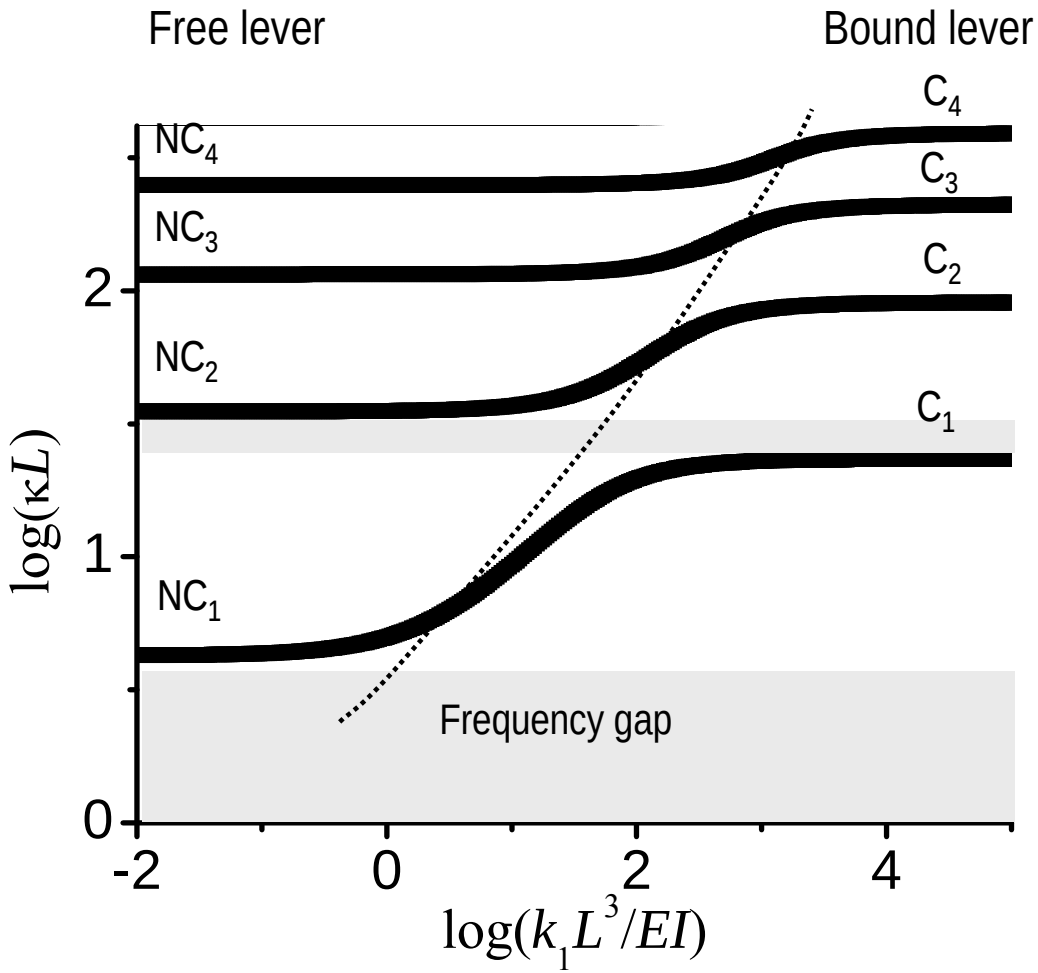

Resonance enhancement in PFM

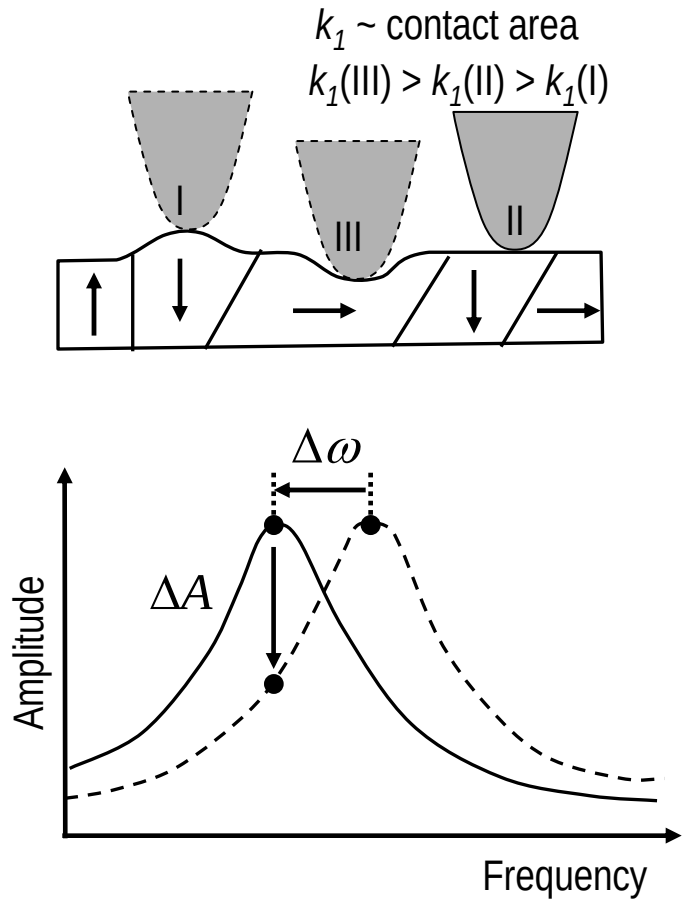
- Away from the resonance: low signal
- At the resonance: strong cross-talk
- Vicinity of the resonance: optimal

If the frequency is too high:

- Cantilever does not respond
- Photodiode bandwidth

# Resonance Enhancement in PFM

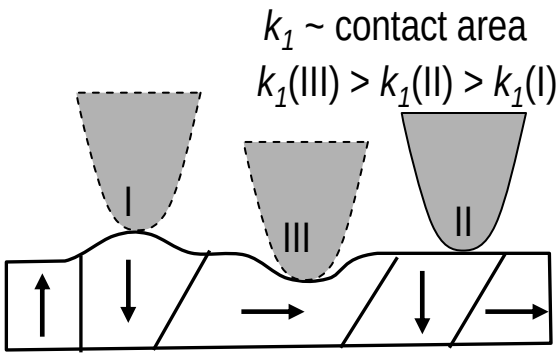


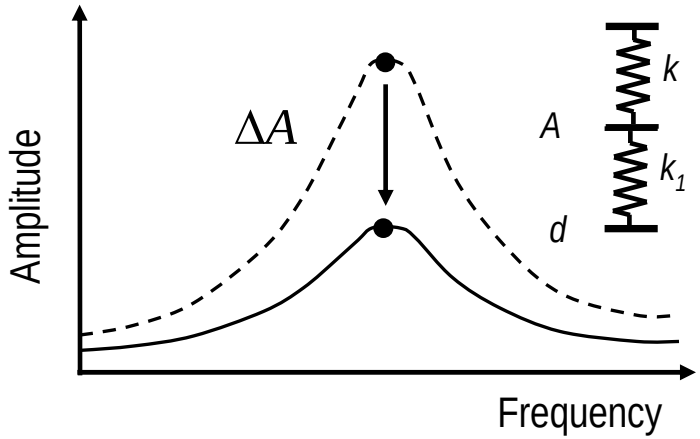




We expect significant variability of resonance frequency as a function of topography!

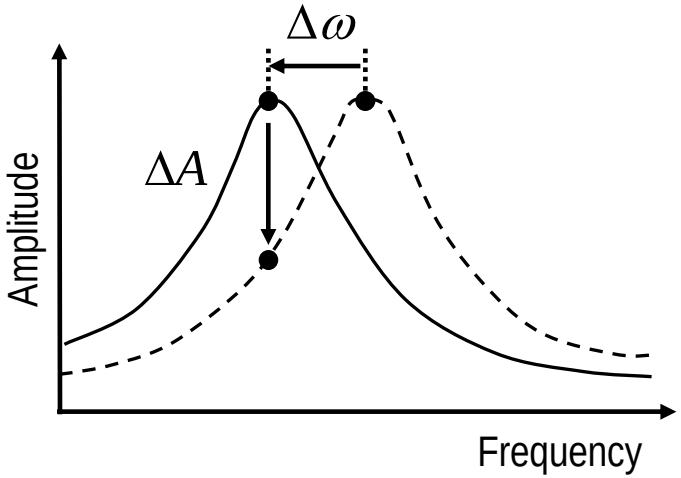
# Resonance Enhancement in PFM

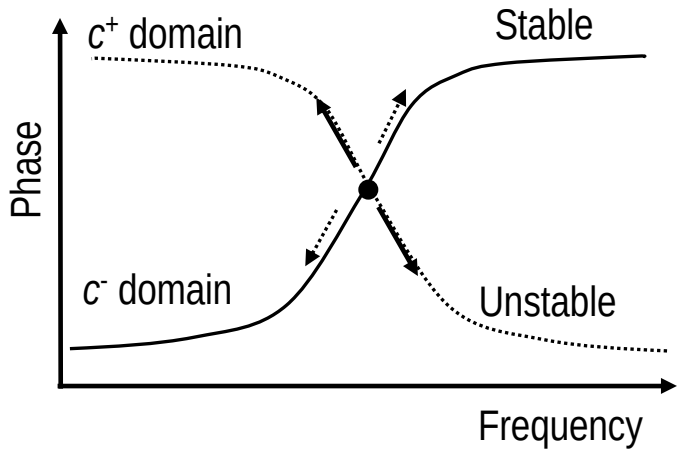
To image weakly piezoelectric materials or high resolution spectroscopy, we need to detect small (1-10 pm) surface displacements. Can we use resonant enhancement?



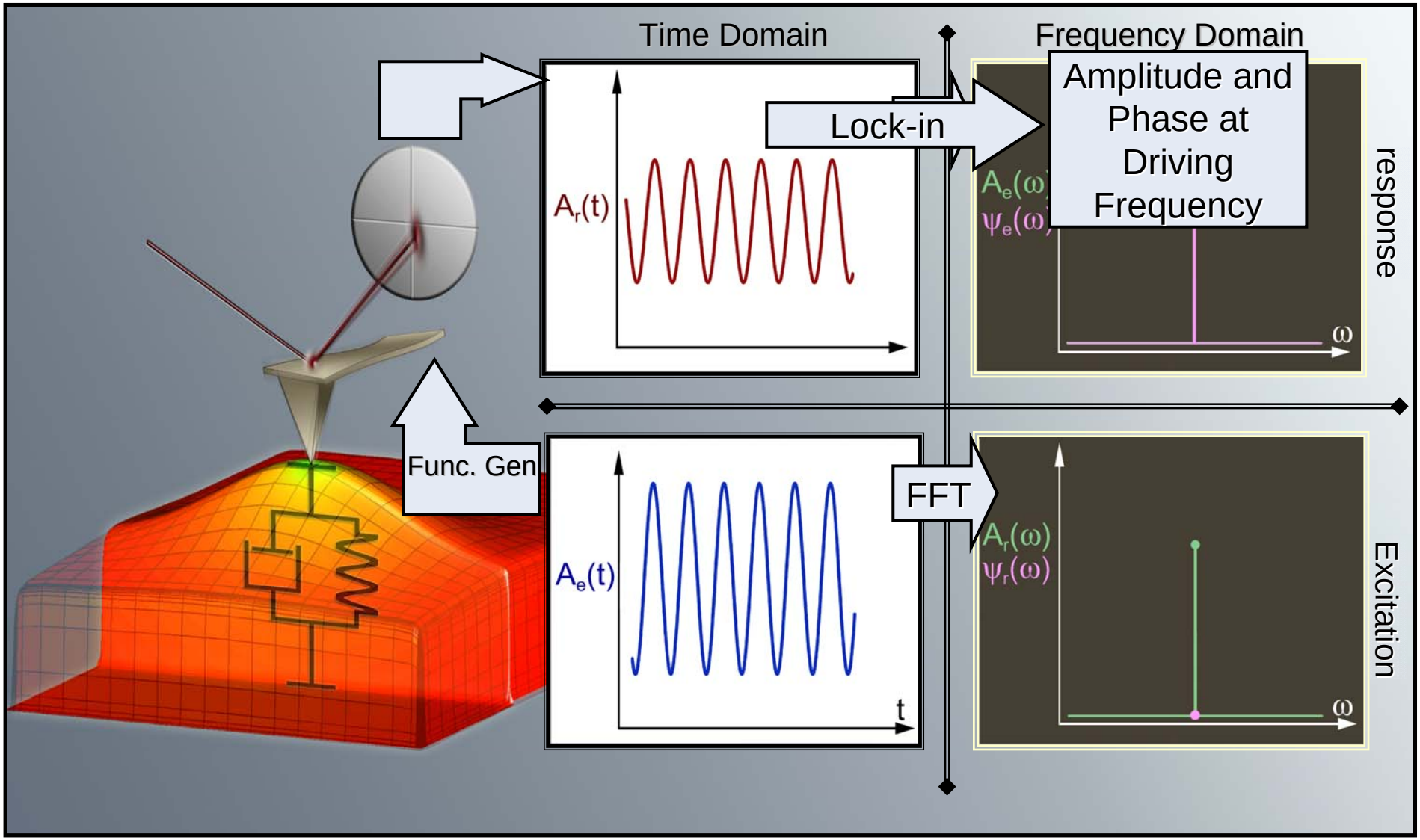

**Problem 1:** resonant frequency depends on topography, and not on electromechanical response!
**Problem 2:** Standard PLL frequency-tracking loops are unstable



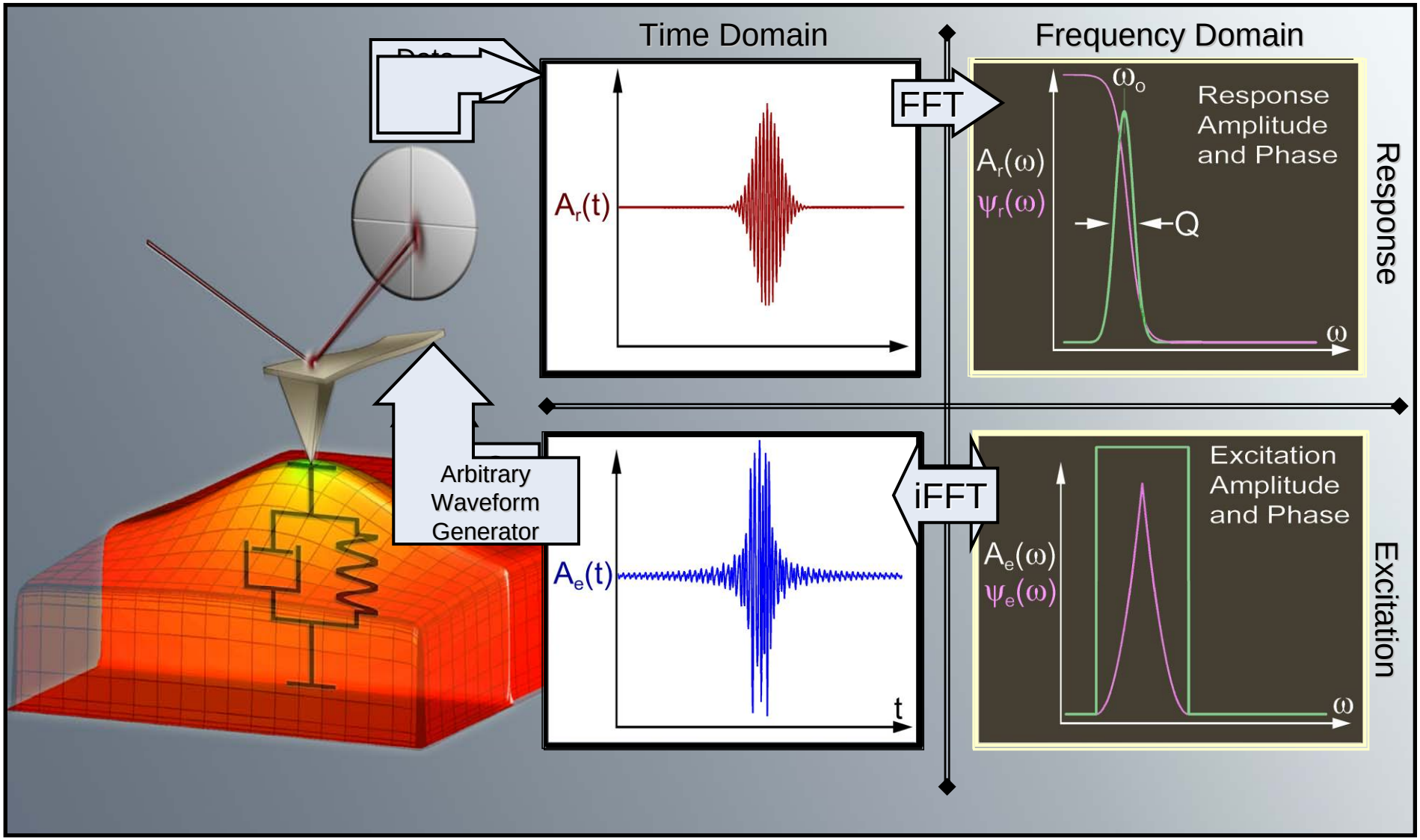




Solution: fast acquisition of amplitude-frequency curve in the vicinity of resonance

# SPM Methods to Date



# The Band Excitation Method

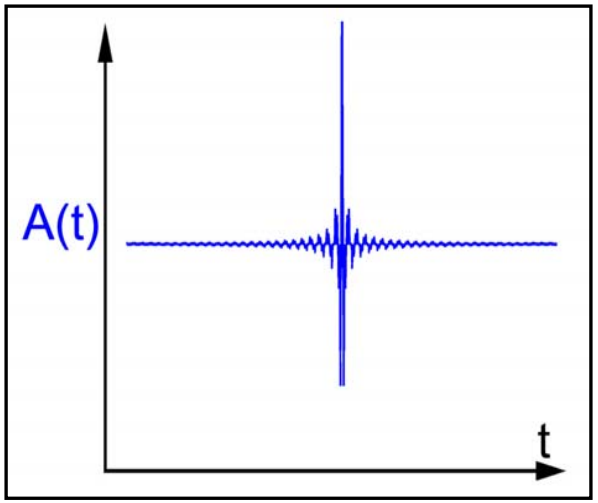


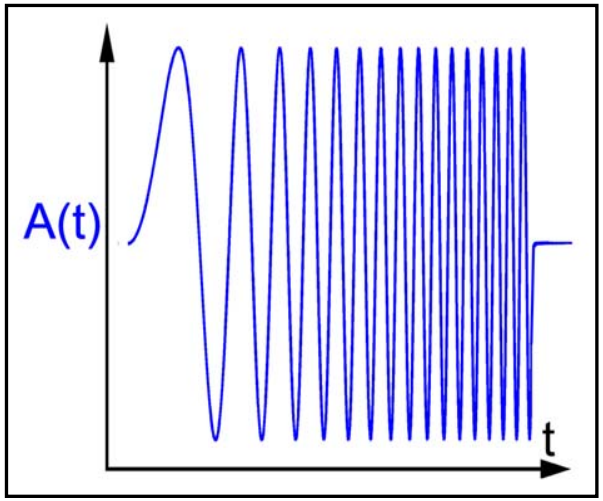
# To $\phi$ or not to $\phi$: The Role of Phase

$$SINC = \frac{\sin(a \cdot t)}{t}$$

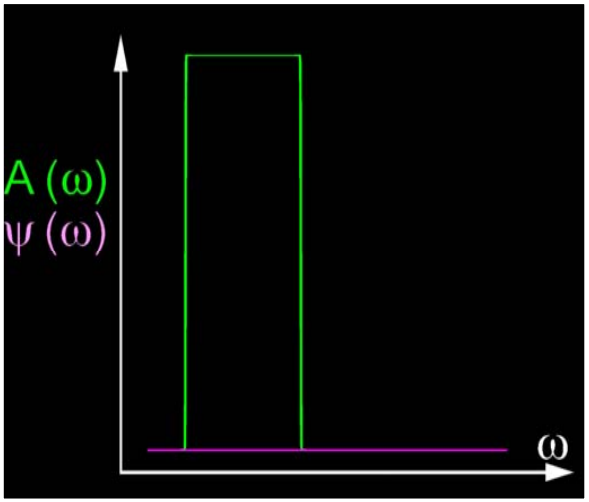
$$CHIRP = \sin\left(\frac{a \cdot t^2}{2}\right)$$

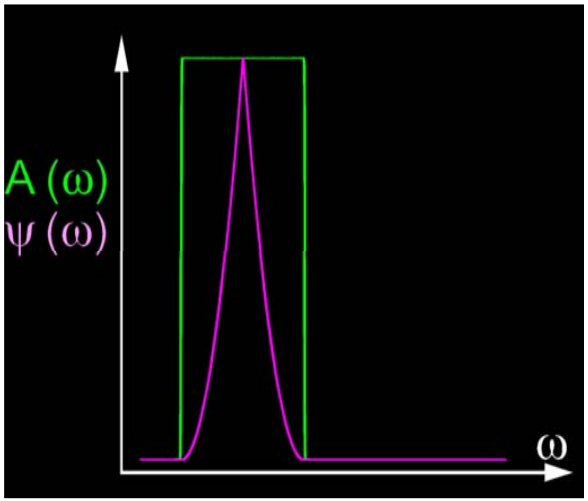
Time Domain




Frequency Domain



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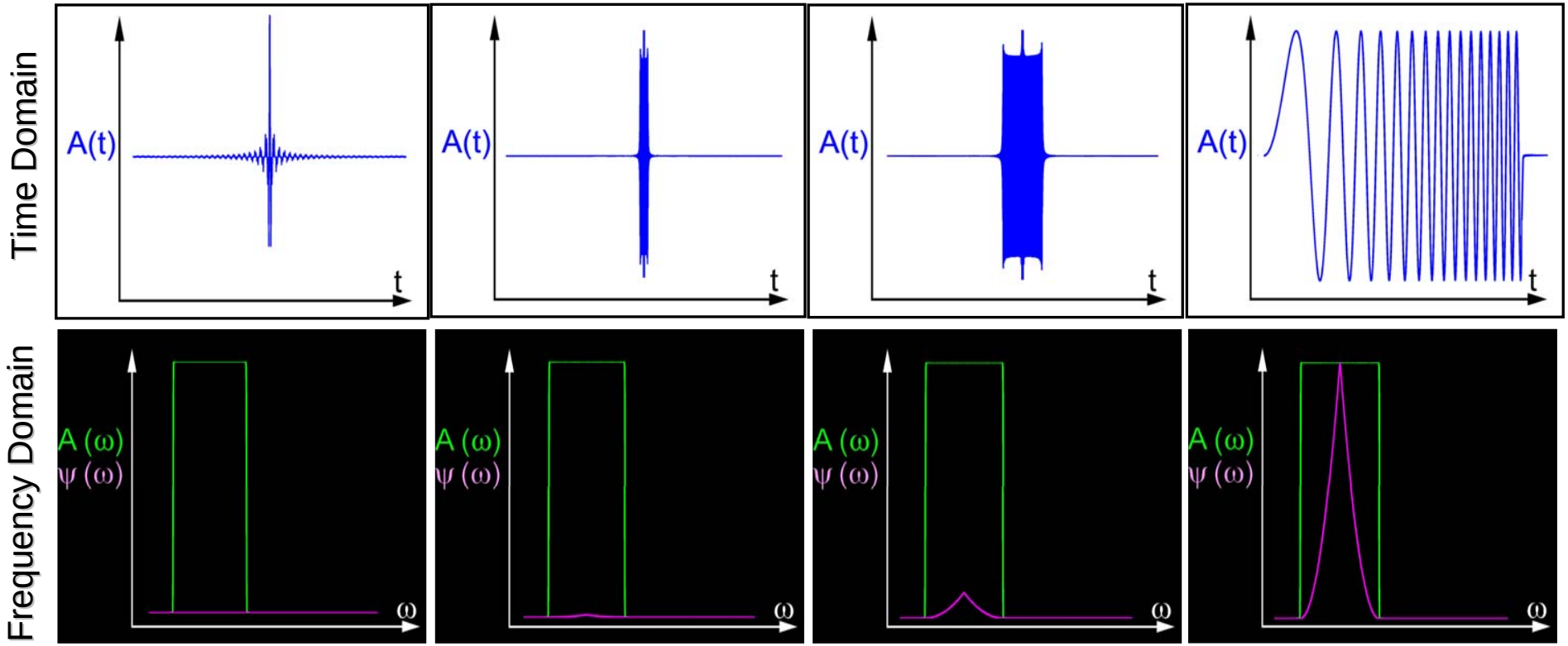
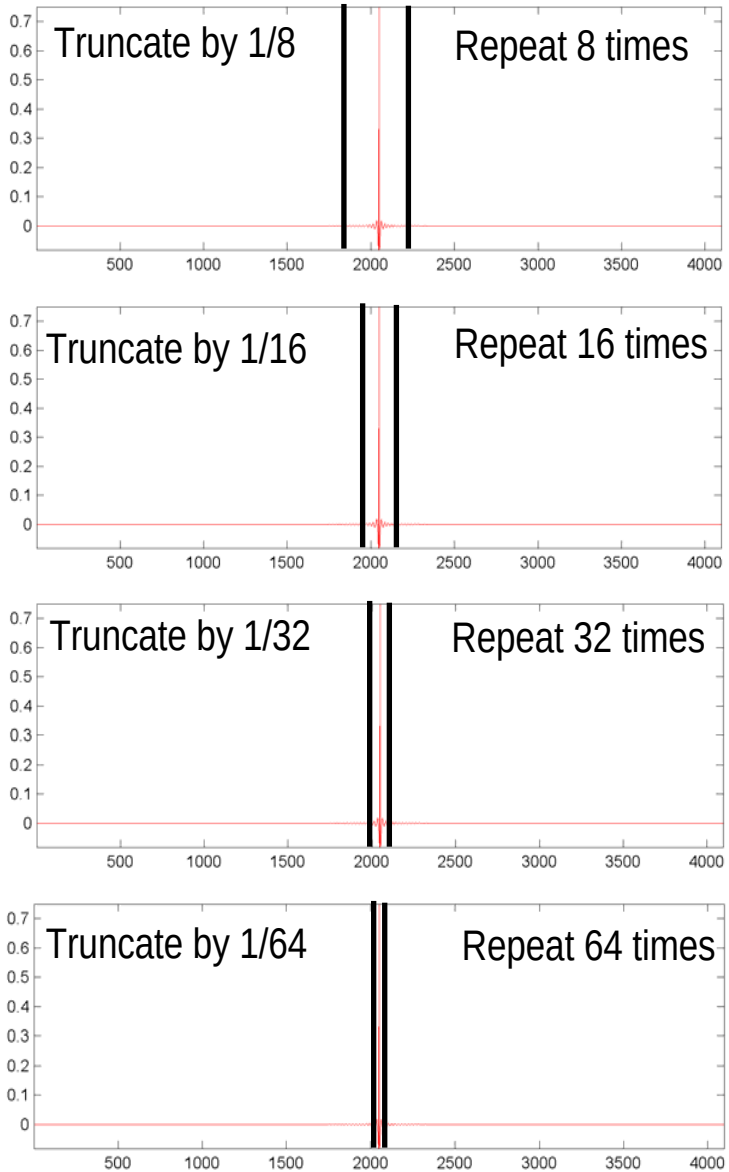
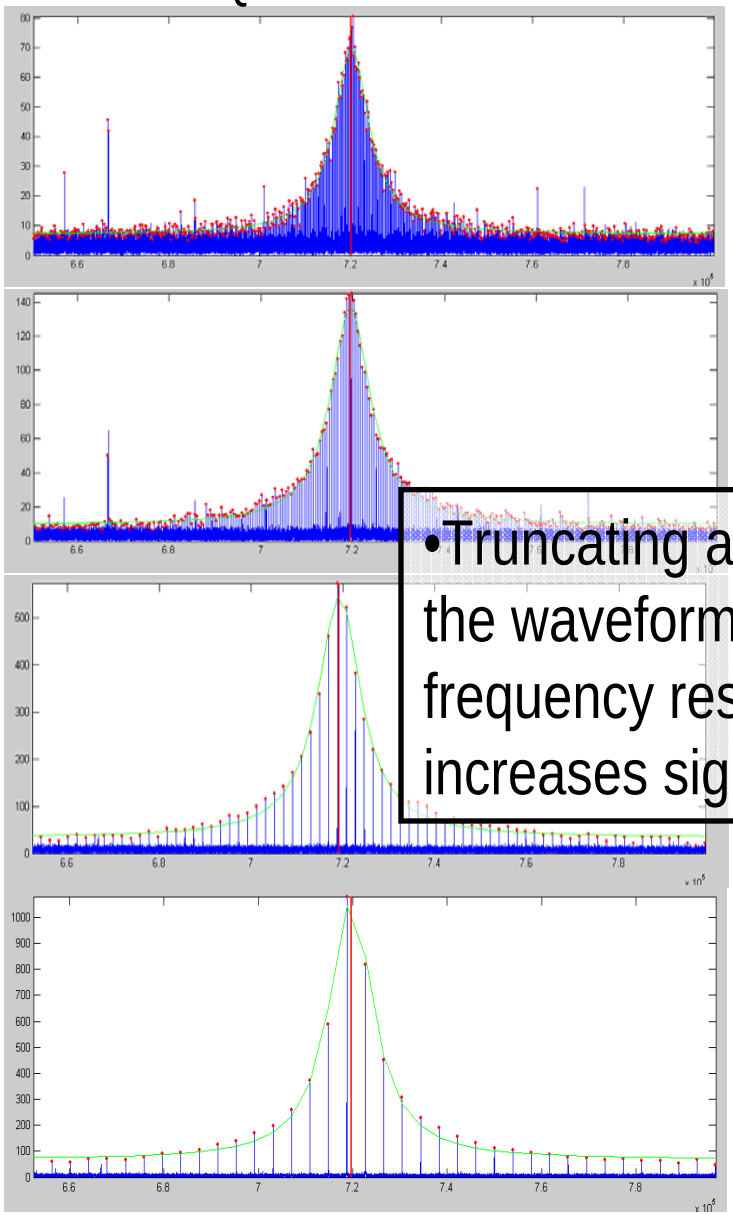


# Truncating the Waveform: Frequency Resolution vs Signal-to-Noise

TIME DOMAIN

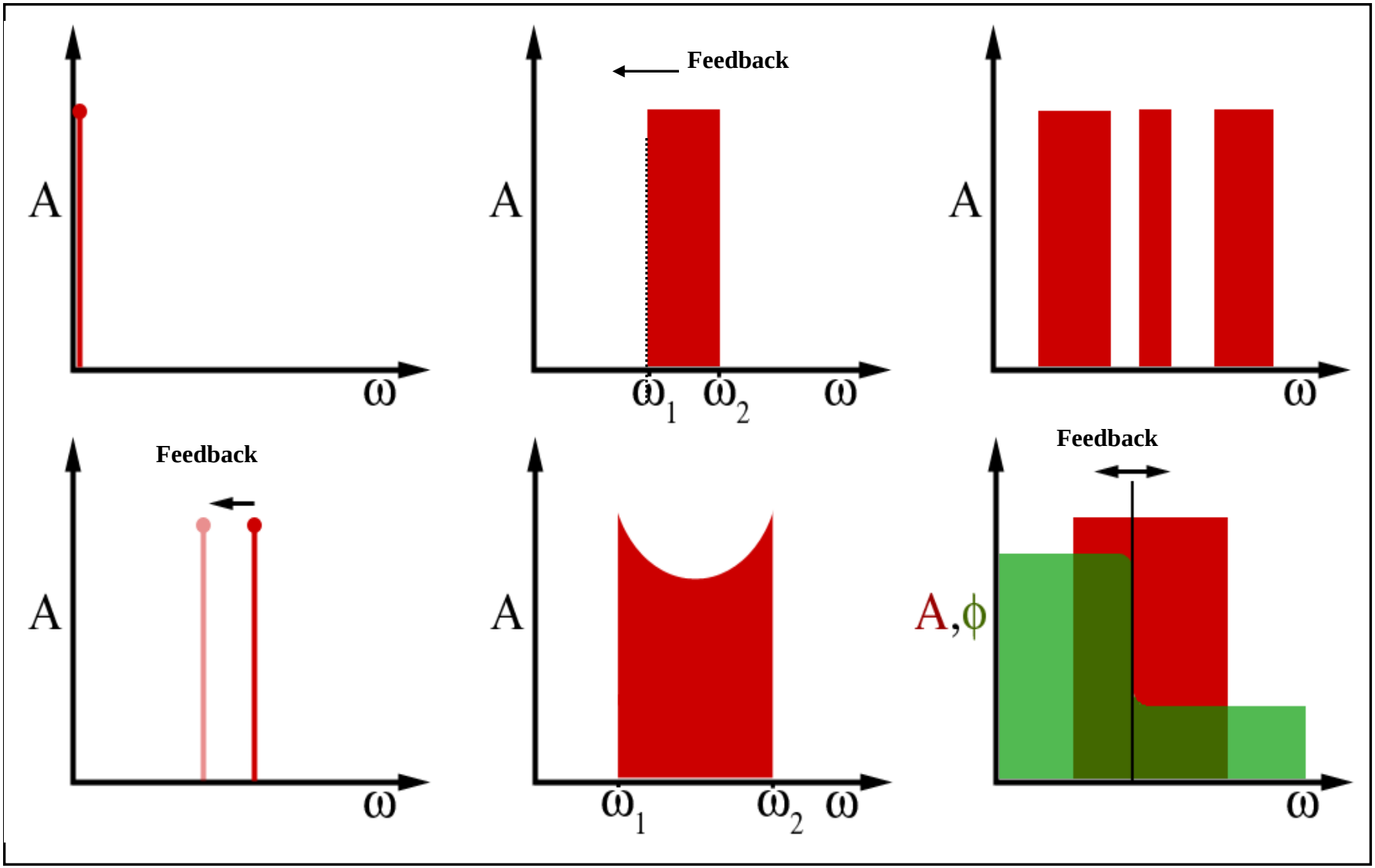



FREQUENCY DOMAIN



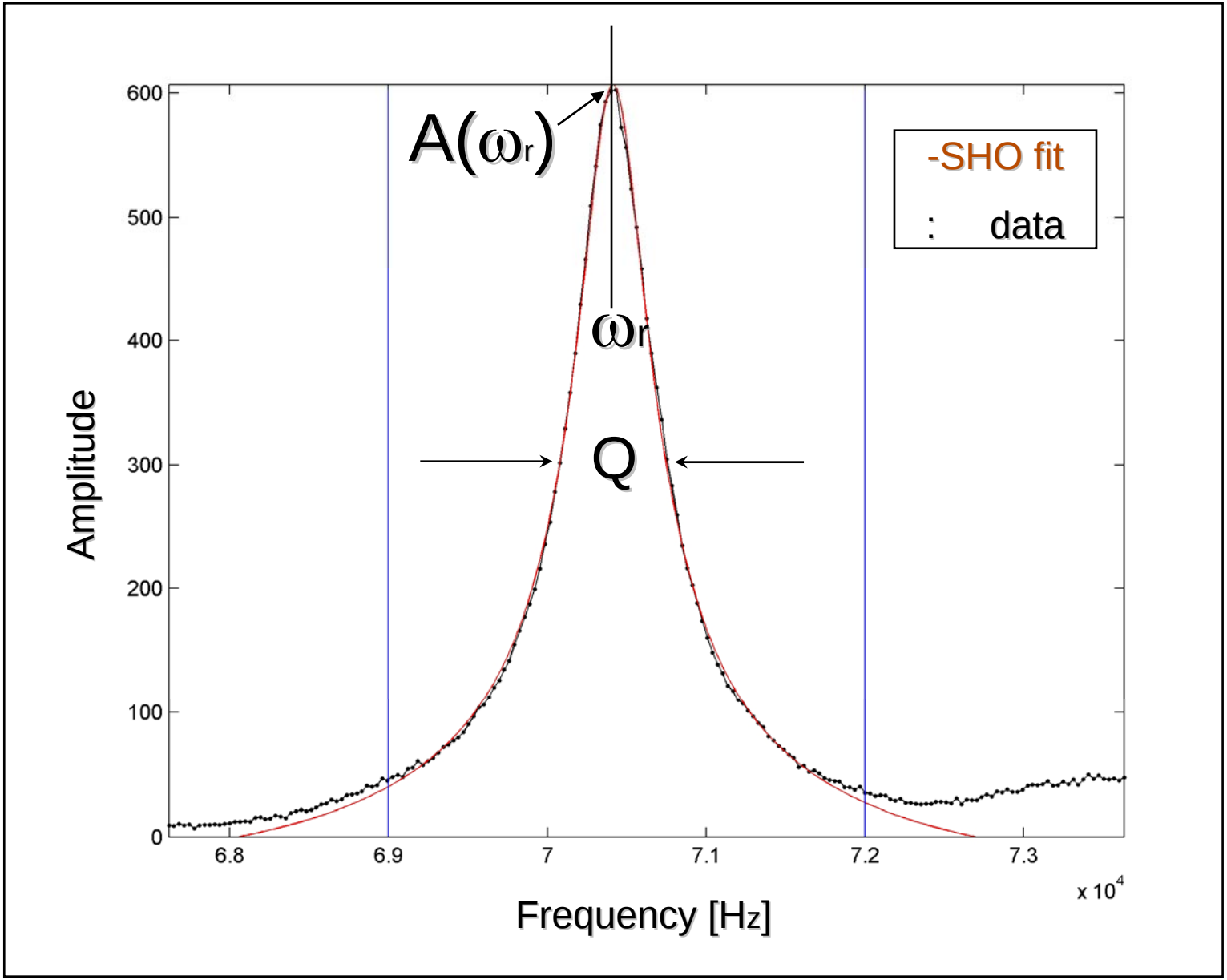
- Truncating and repeating the waveform reduces the frequency resolution but increases signal-to-noise

# Advanced BE Options



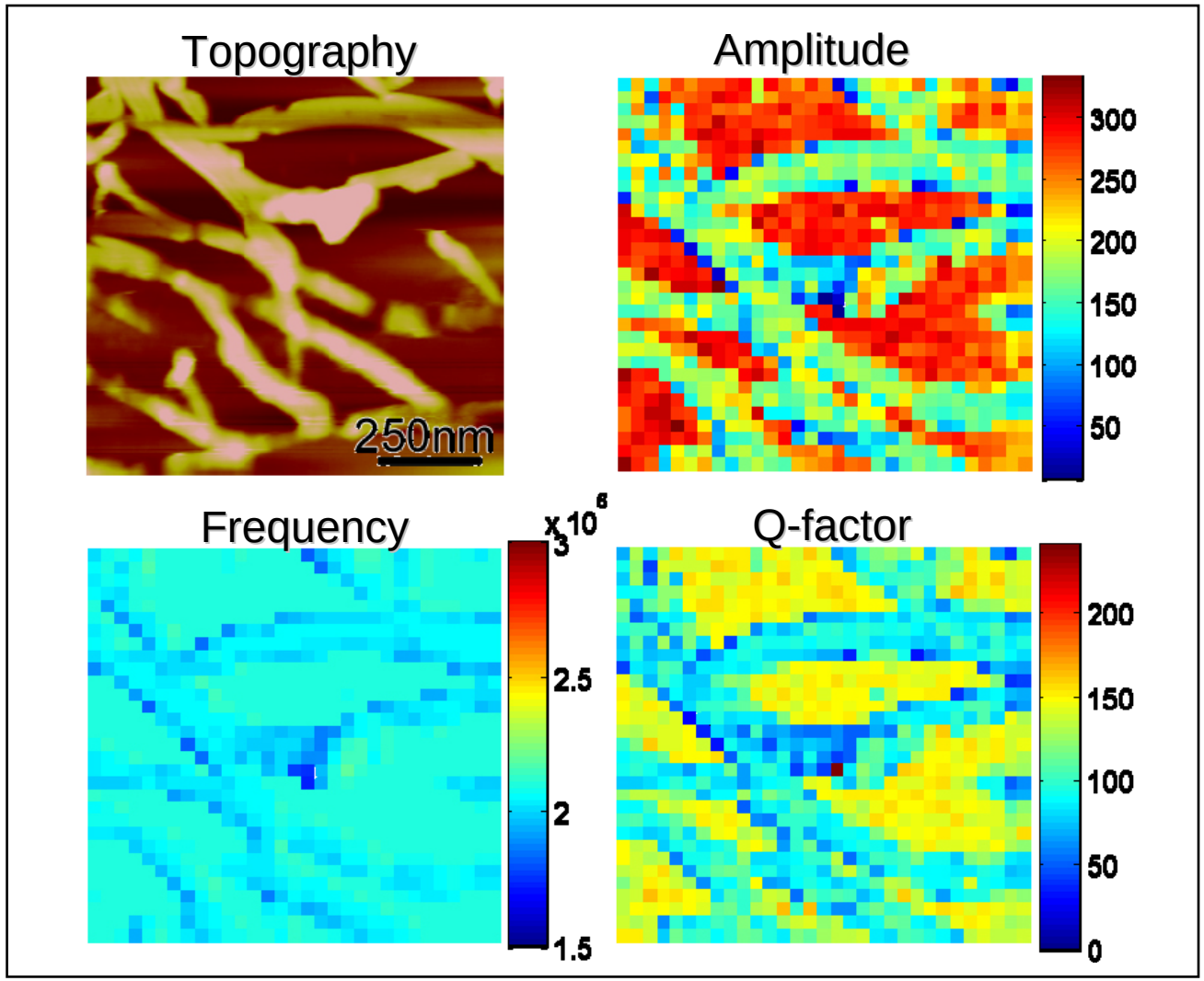

Flexibility to tune excitation and detection windows, sample tails of resonance peak, implement fully digital Q-control, etc.

# Band Excitation in Action



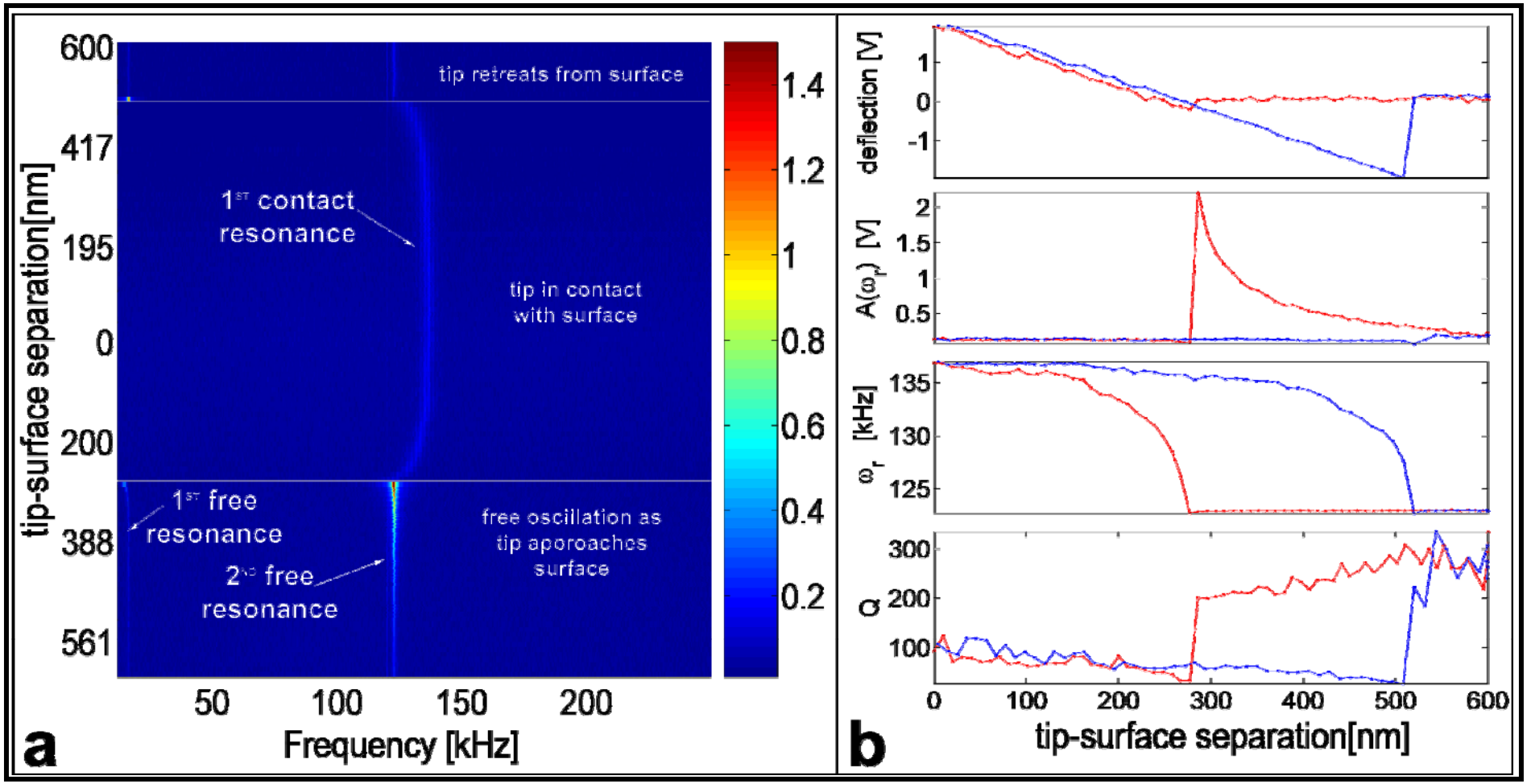

With BE, we collect full response curve at each pixel at standard imaging rate

# Atomic Force Acoustic Microscopy



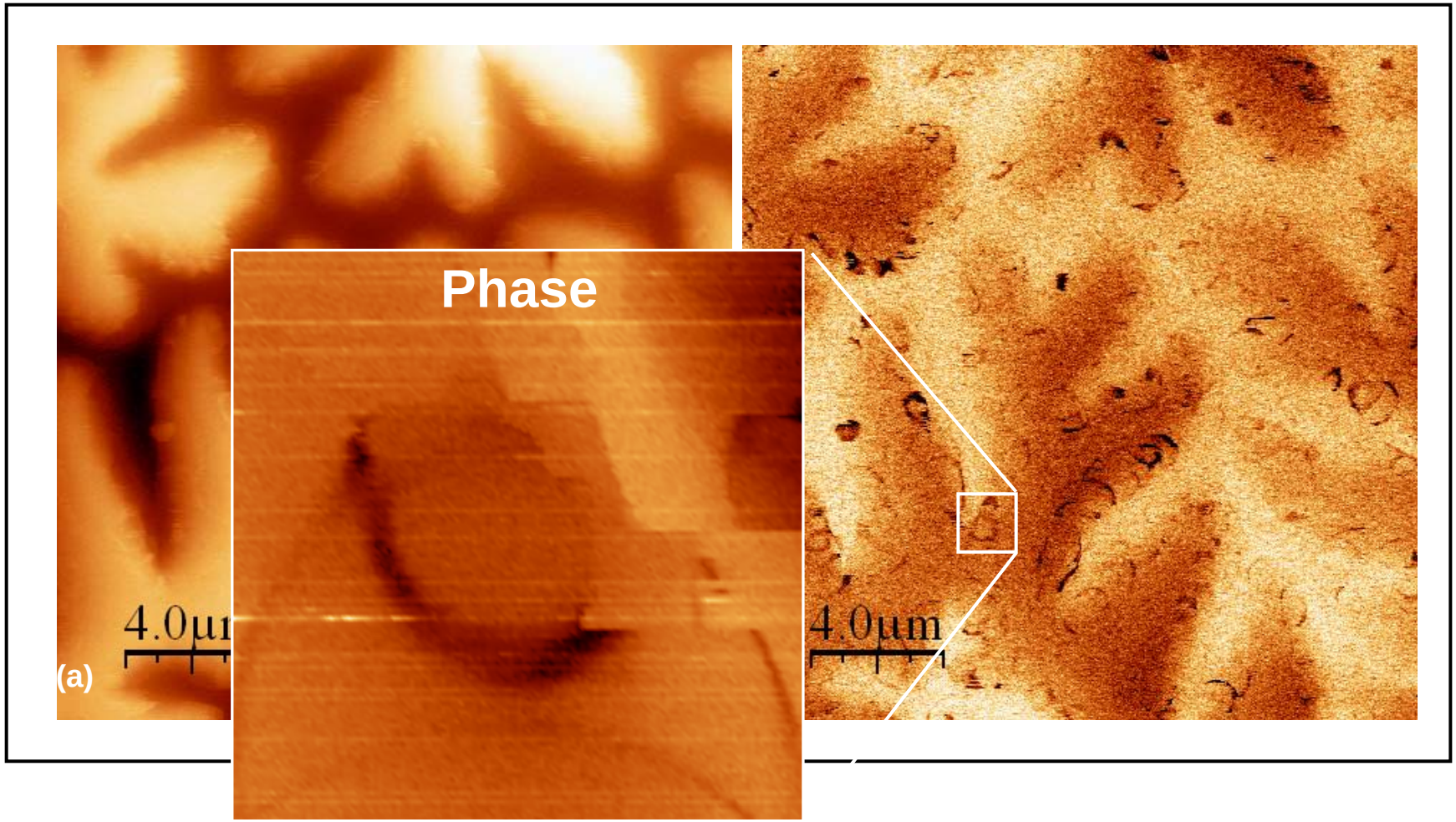

Tobacco mosaic virus (A. Balandin, UCR)

# Force-Distance Spectroscopy



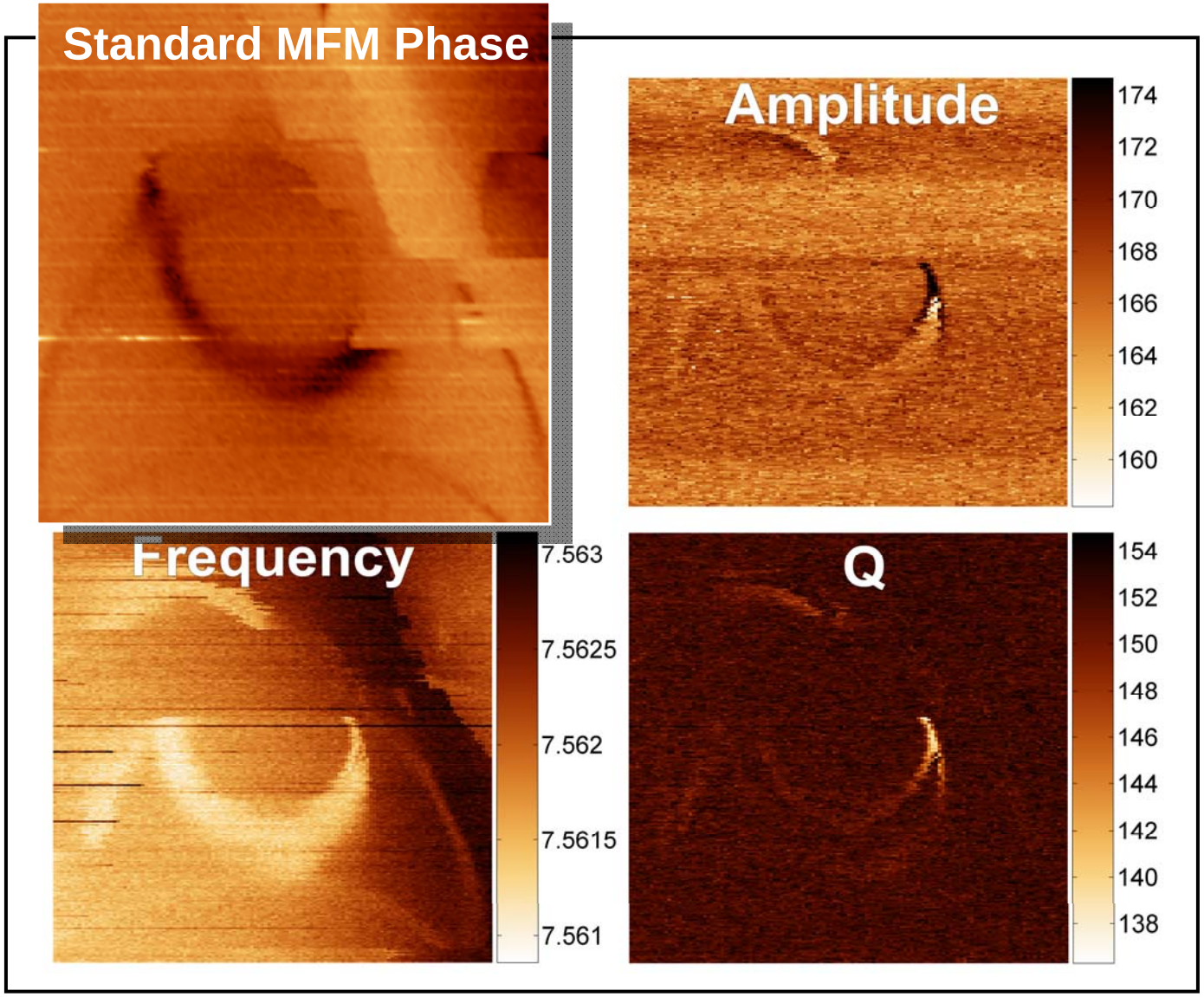

Mica in air

# Magnetic Force Microscopy




Single Crystal Yttrium-Iron Garnet (sample courtesy of R. Proksch, Asylum Research)

# Magnetic Force Microscopy



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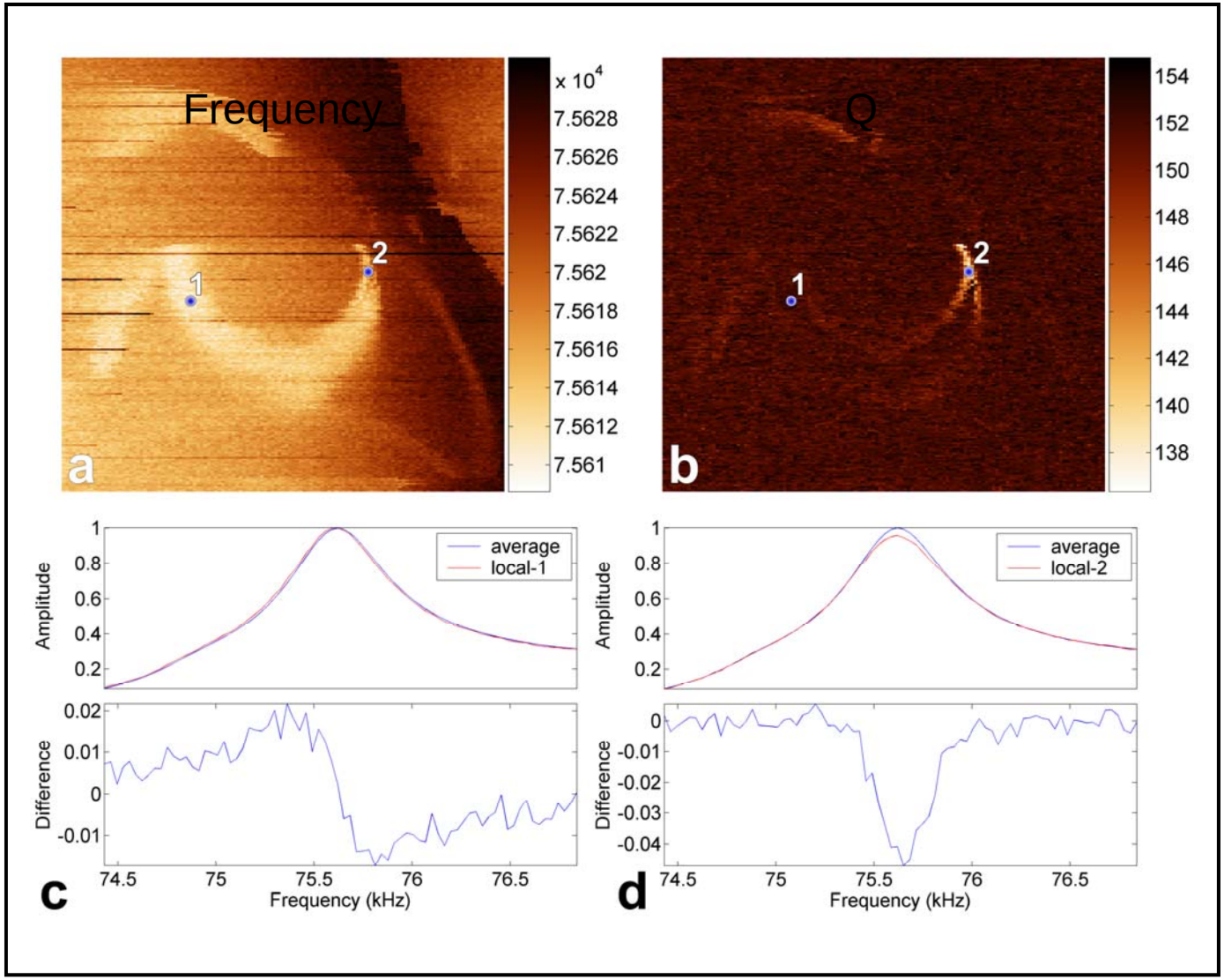
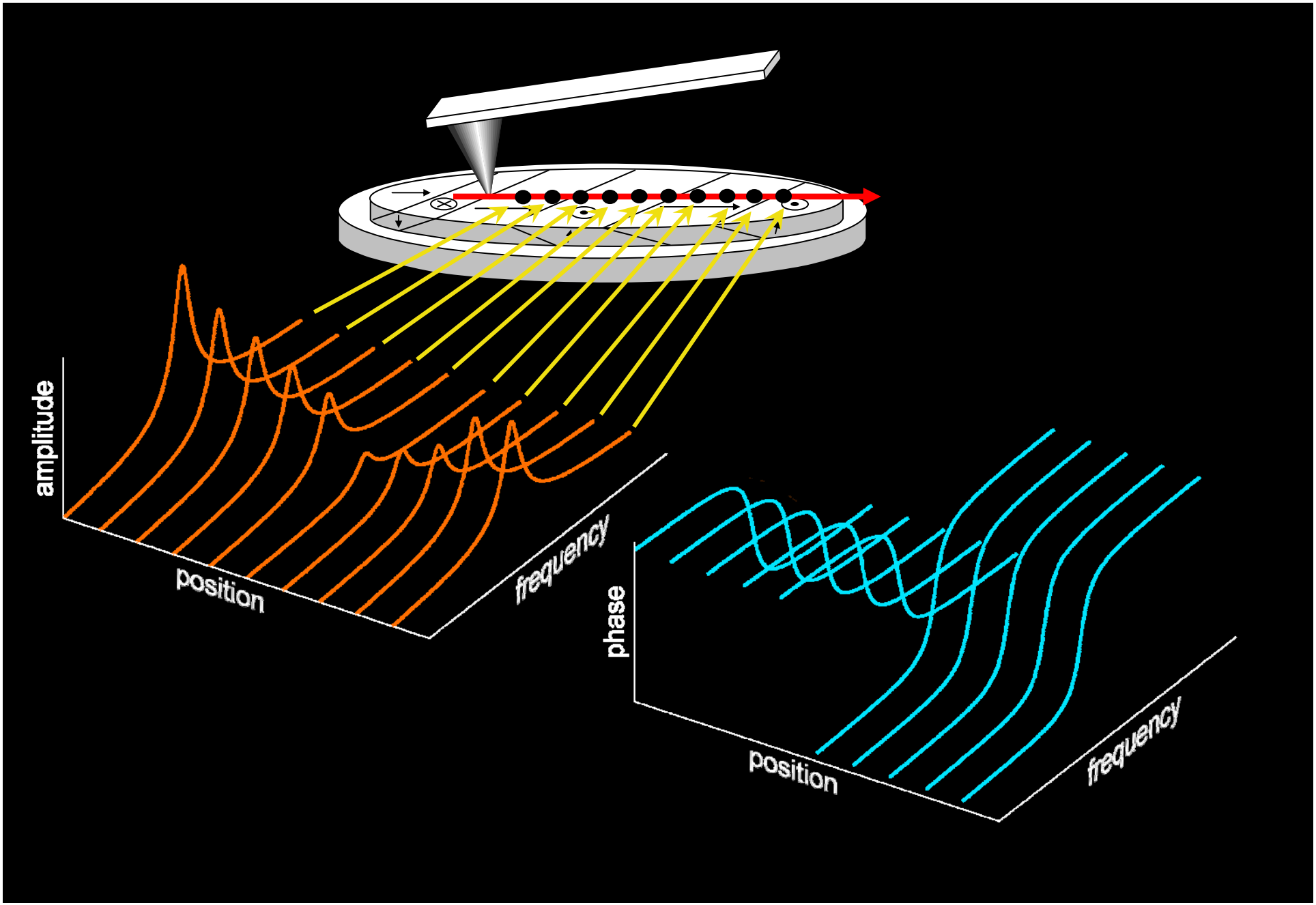



Unambiguous decoupling of conservative and dissipative interactions

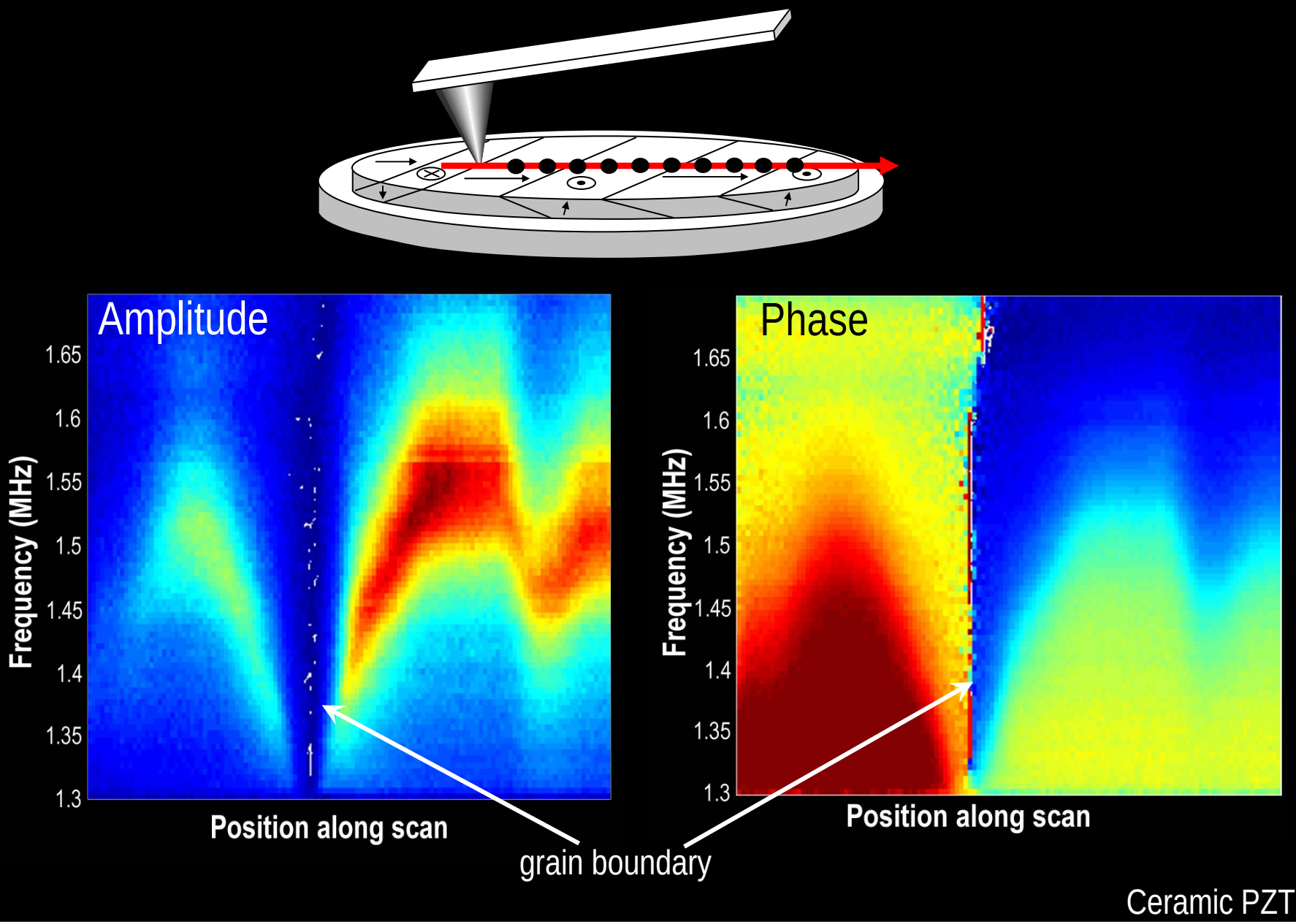
# The Band Excitation Method:

- Provides a more comprehensive description of probe-sample interaction dynamics for a wide range of SPM techniques
- Measures response spectra at every point in a scan
- Fast
- Yields quantitative maps of resonance, amplitude as a function of frequency, and dissipation

# Band Excitation PFM

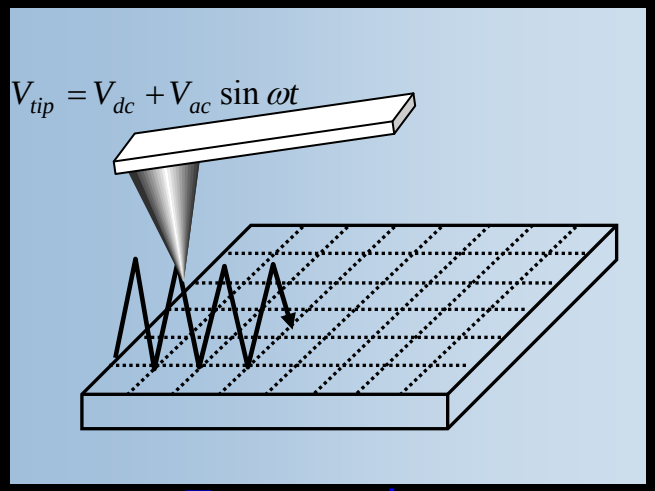


# Band Excitation PFM

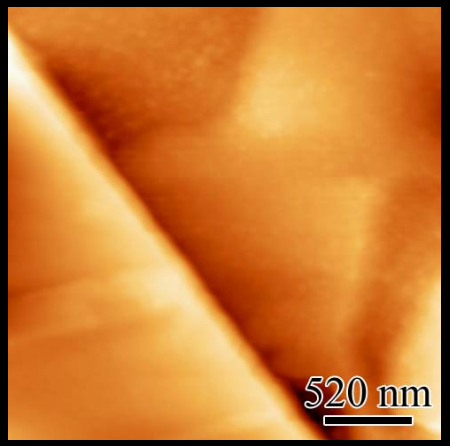


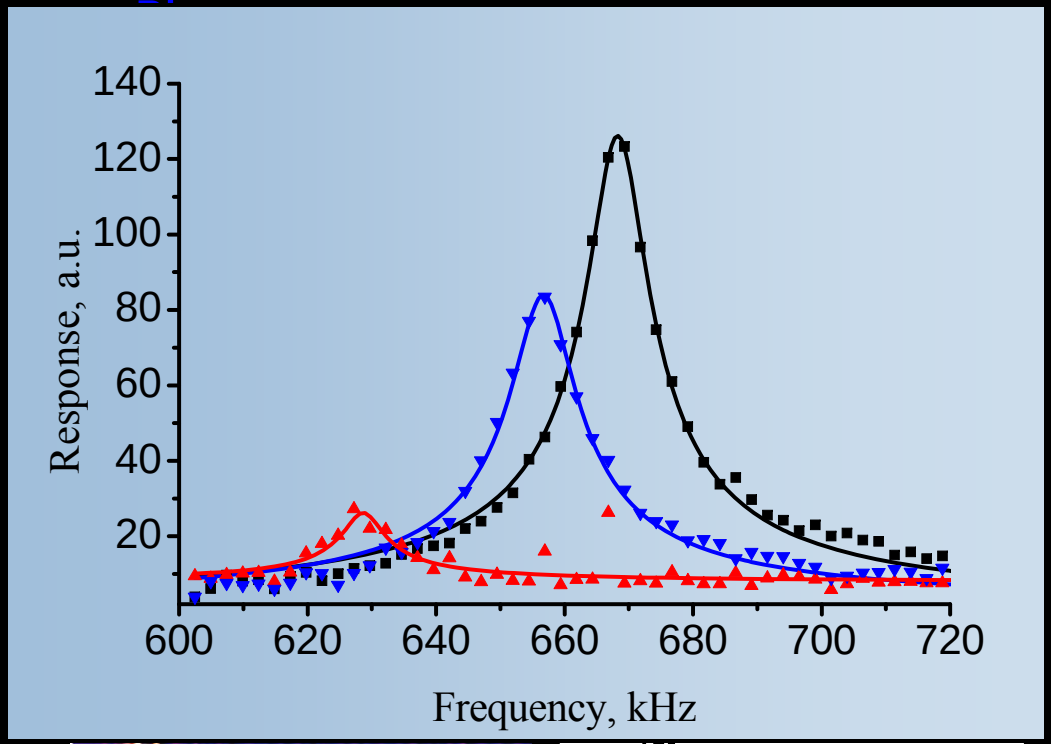

Ceramic PZT

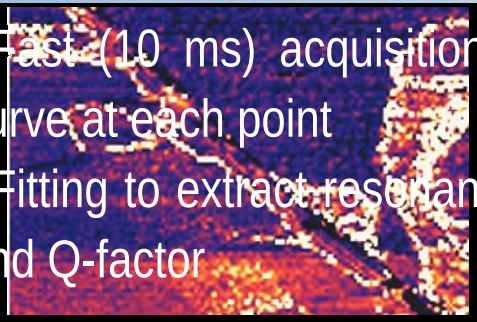
# Band Excitation PFM

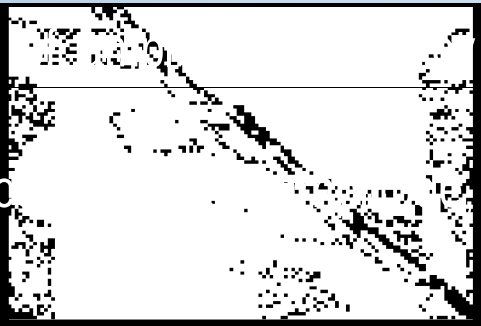



Topography



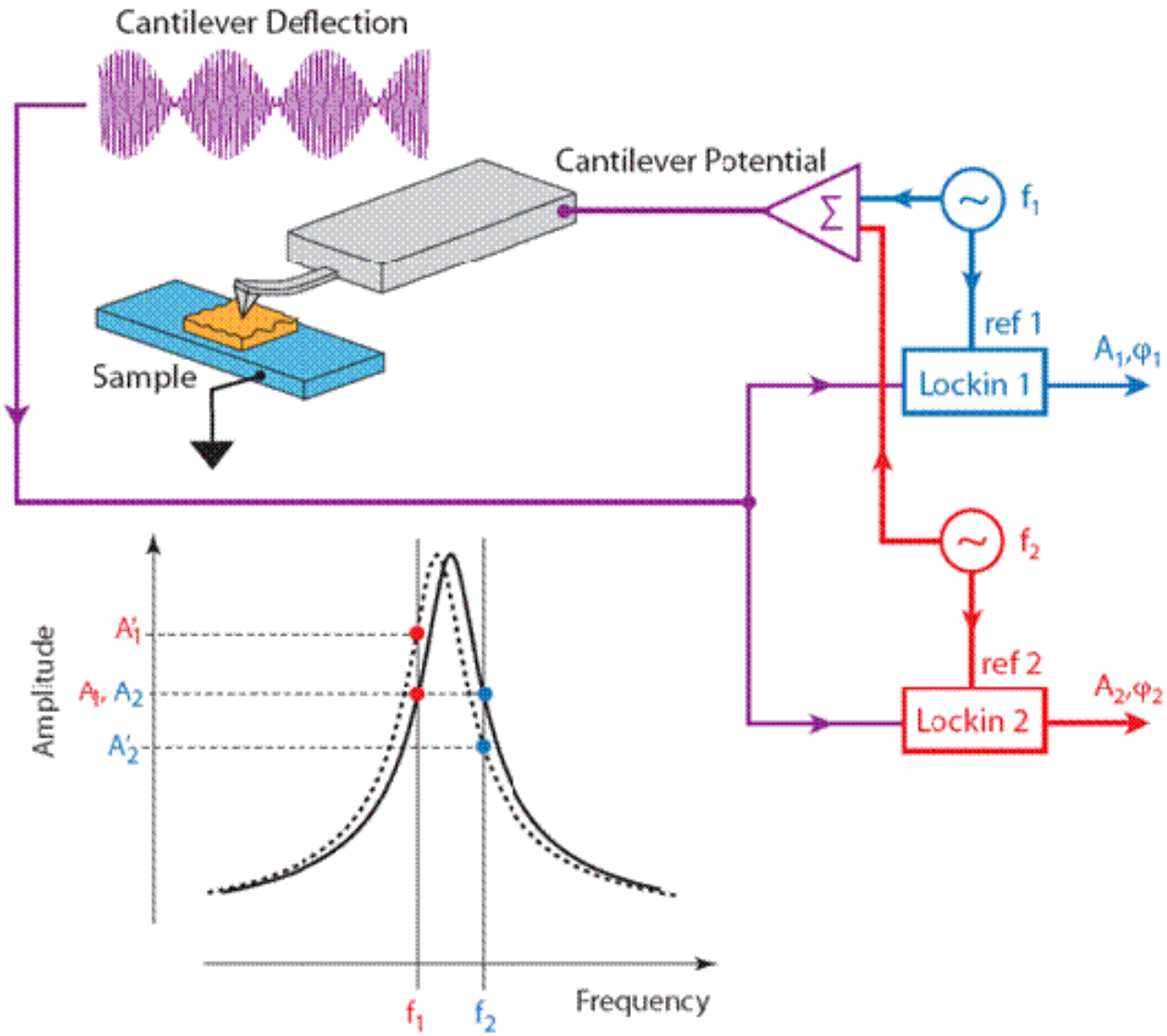





- Fast (10 ms) acquisition curve at each point
- Fitting to extract resonanc and Q-factor

Driving the cantilever electrically, we are able to obtain maps of piezoresponse at the resonance, resonant frequency and Q-factor as we move from point to point on a sample. There are differences in electromechanical and elastic properties.

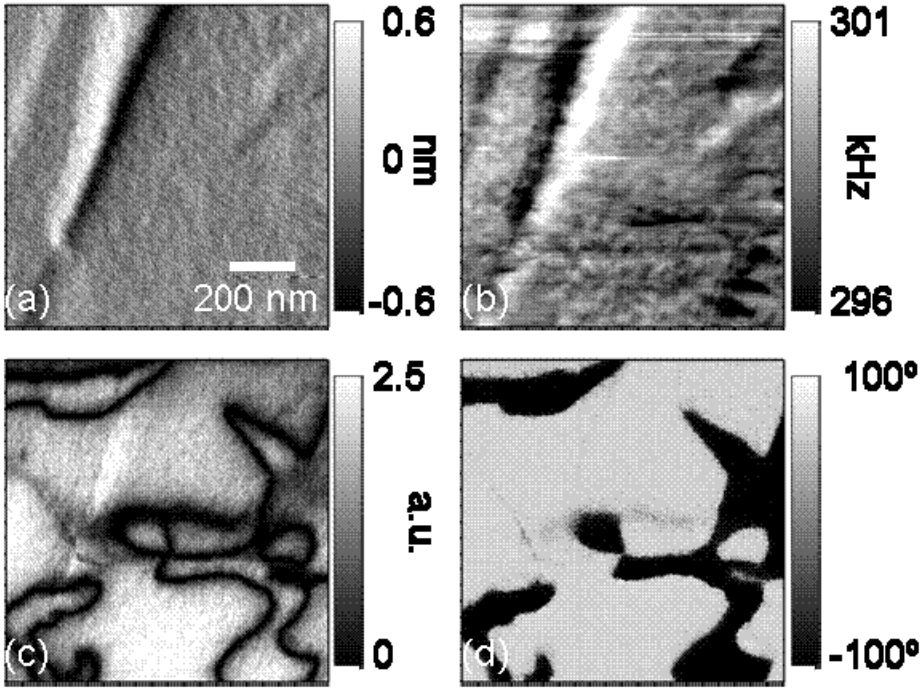
# Dual-Frequency Resonance Tracking PFM



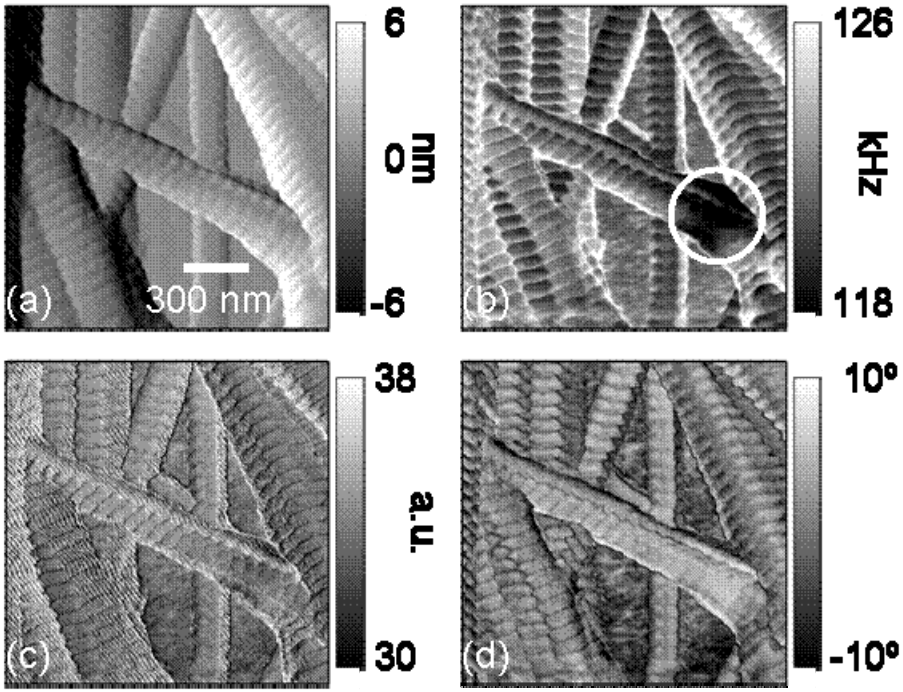

Amplitude based resonance frequency feedback

# Dual Frequency Resonance Tracking PFM

Tooth dentine

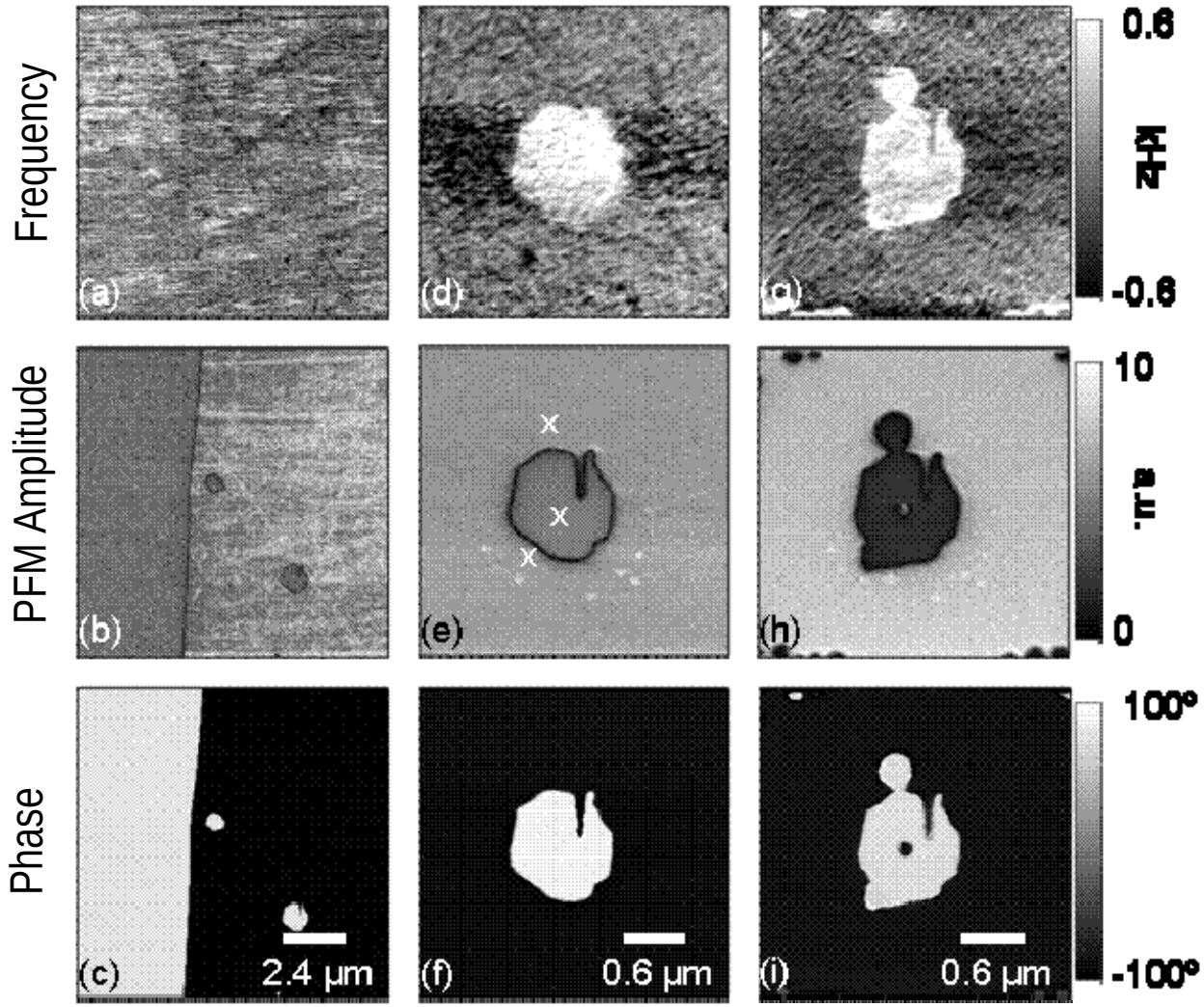



Collagen fibril



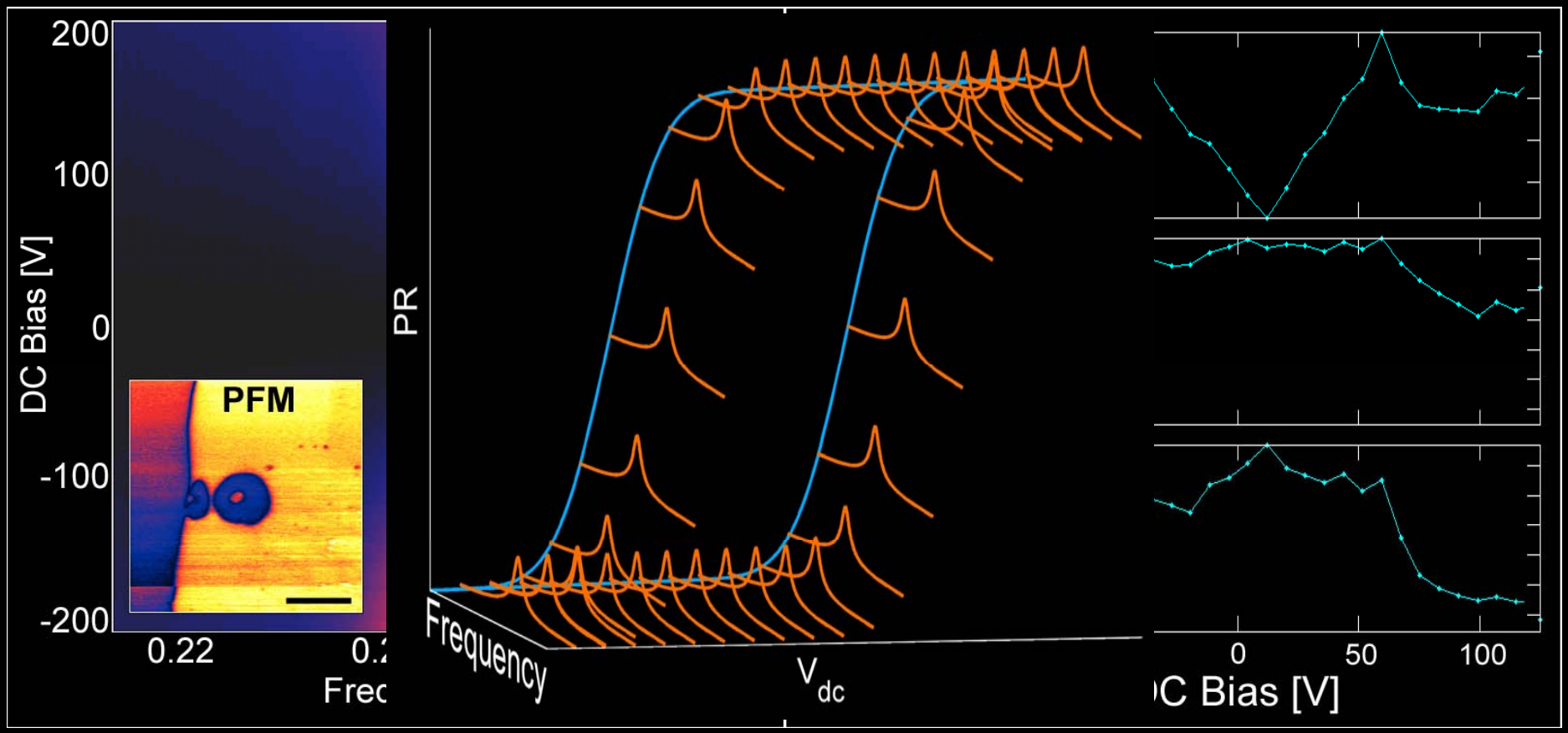

Topography, piezoresponse, and resonance frequency are now decoupled

# Dual Frequency Resonance Tracking PFM



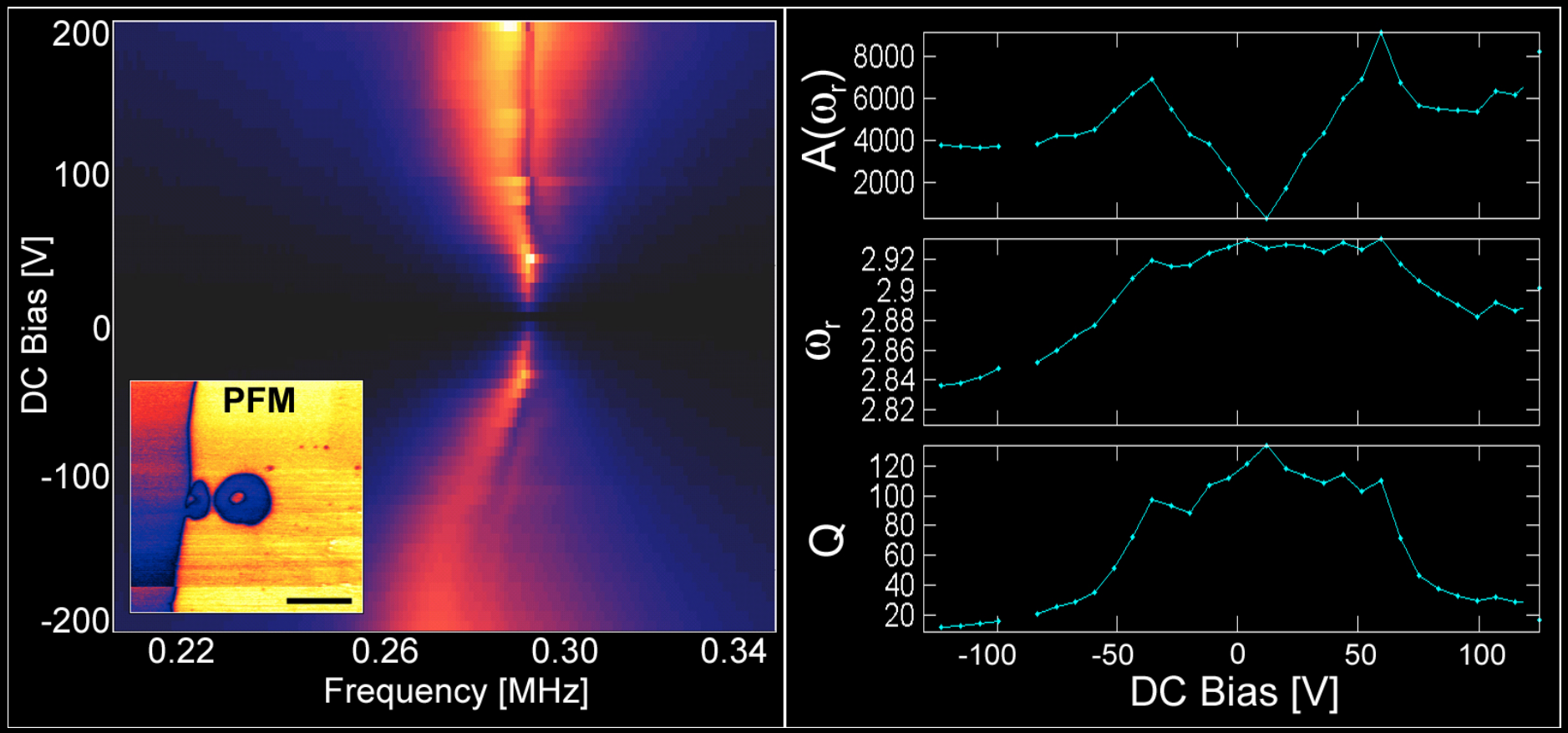

Sometimes, we observe resonance frequency dependence on polarization even for flat surfaces!

# Band Excitation PFS



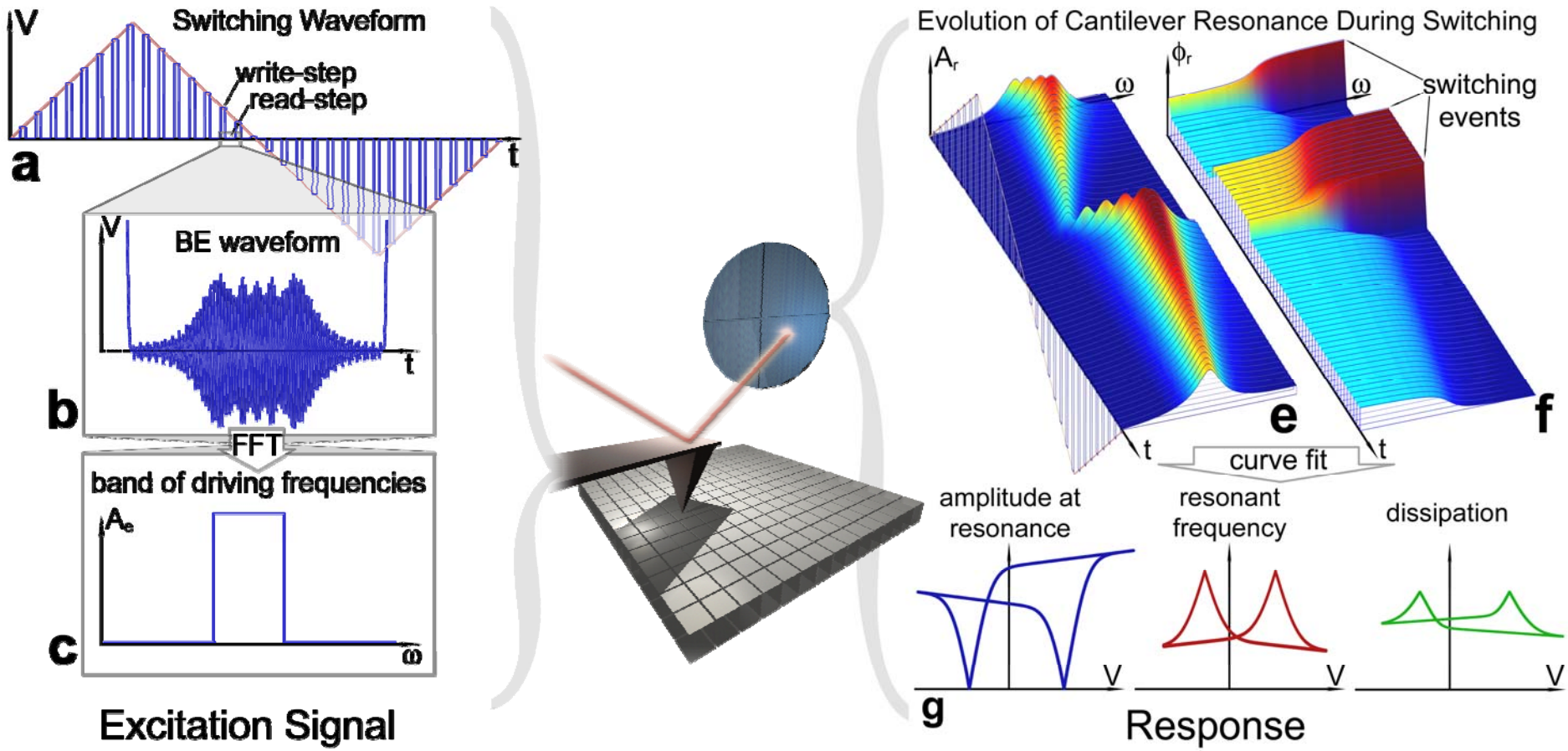

Periodically Poled Lithium Niobate

# Band Excitation PFS




Systematic evolution of resonance frequency and dissipation along the hysteresis loop.

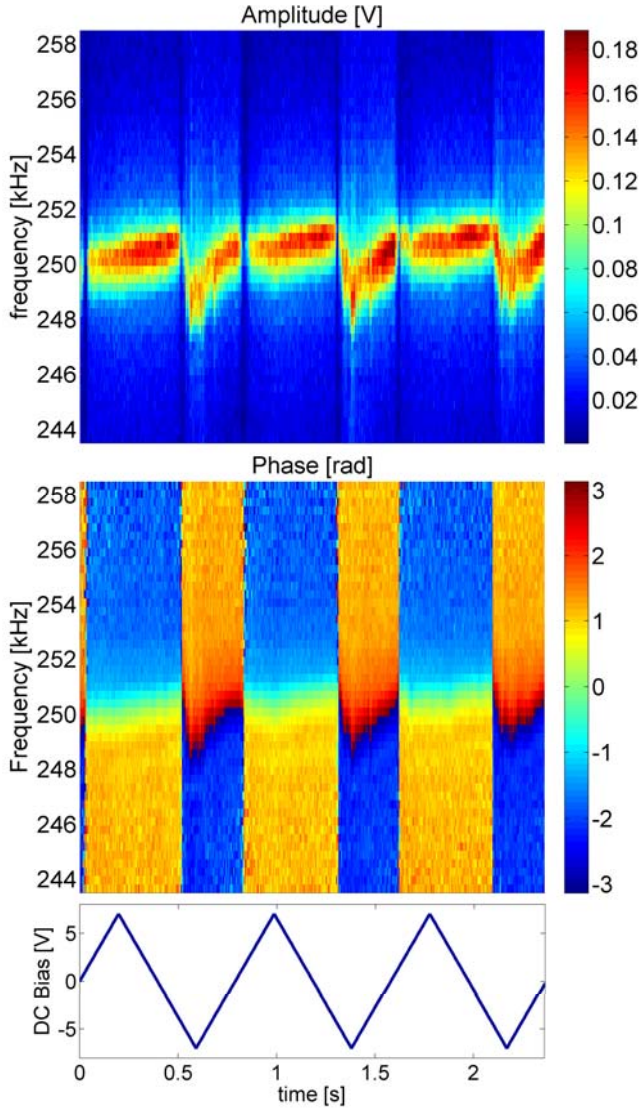
# Band Excitation SS-PFM

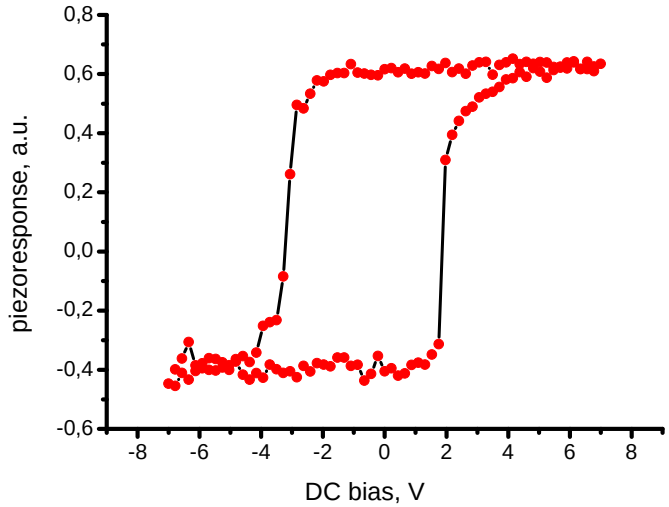


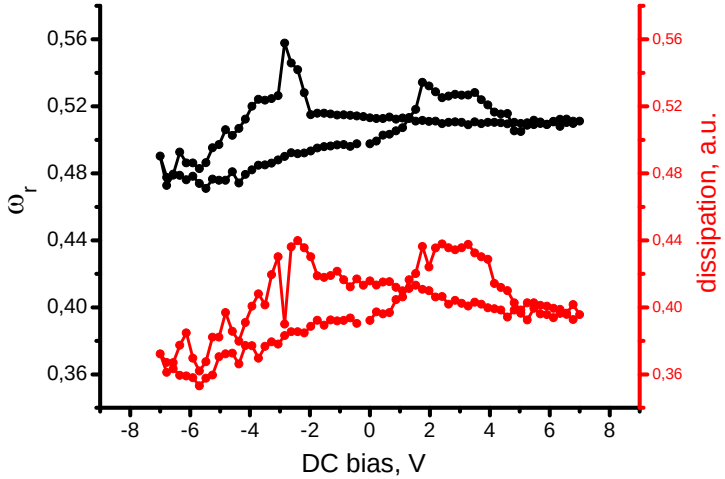

Typical 4D data array size: ~3 GB
Processing time: ~10 hours

Combination of BE and SS-PFM allows systematic studies of voltage-dependence mechanics and polarization switching on surfaces

# Band Excitation SS-PFM in Ultra High Vacuum

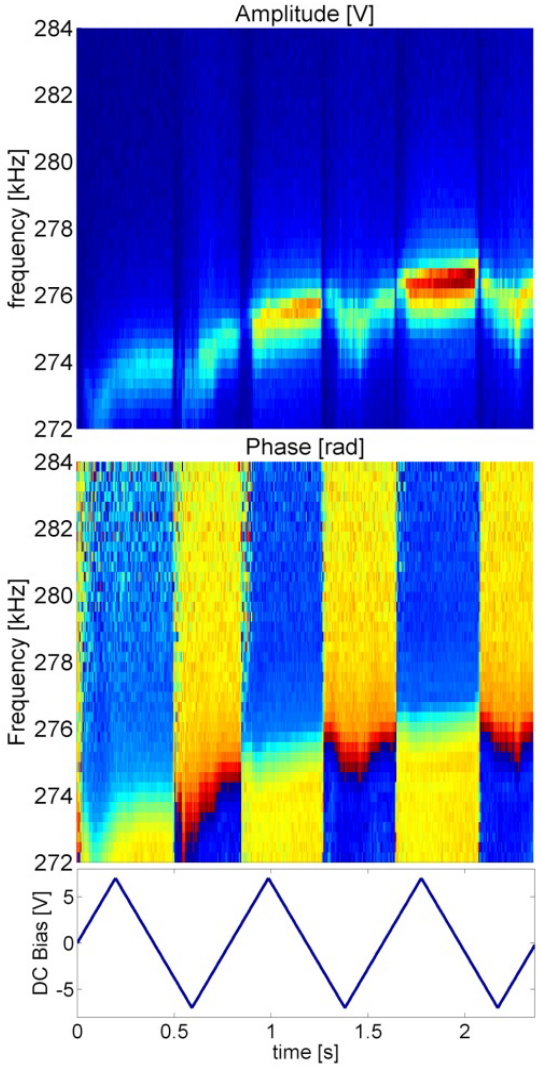


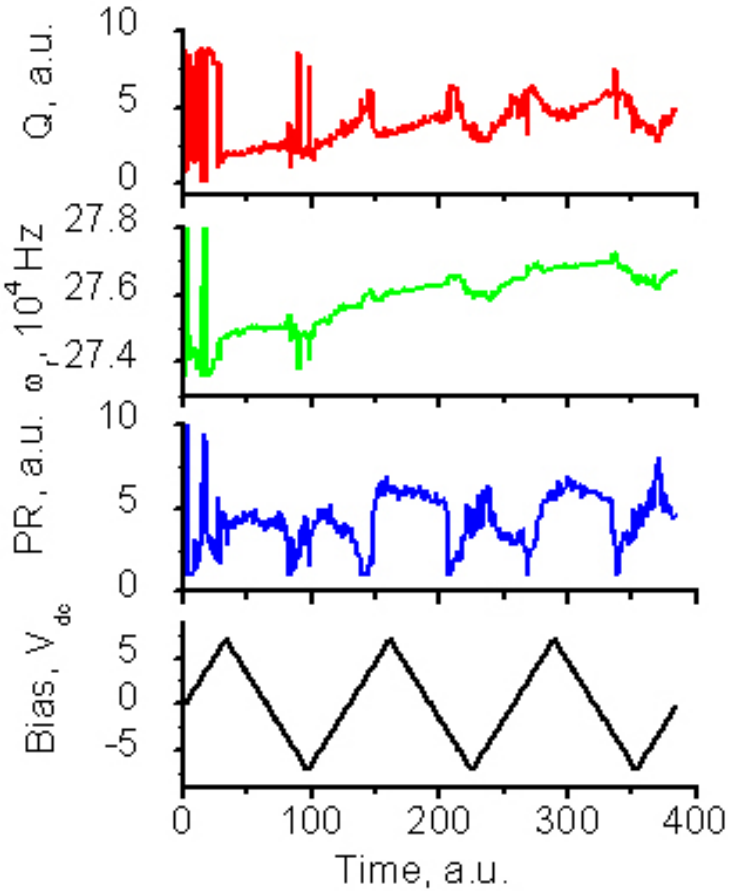




Reproducible changes of contact stiffness during switching

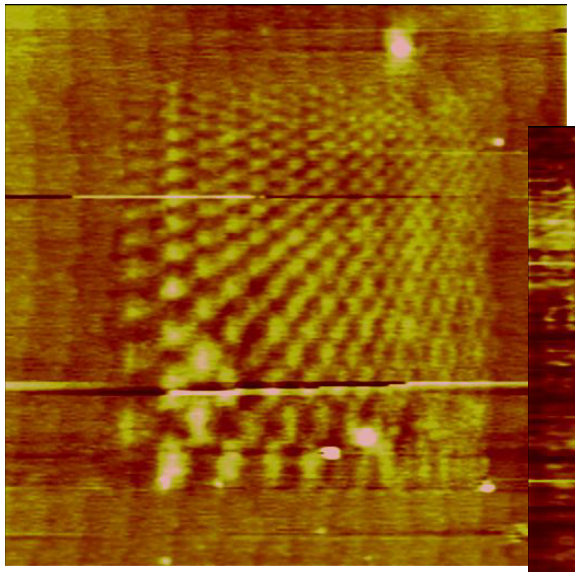
# Band Excitation SS-PFM in Ambient Environment



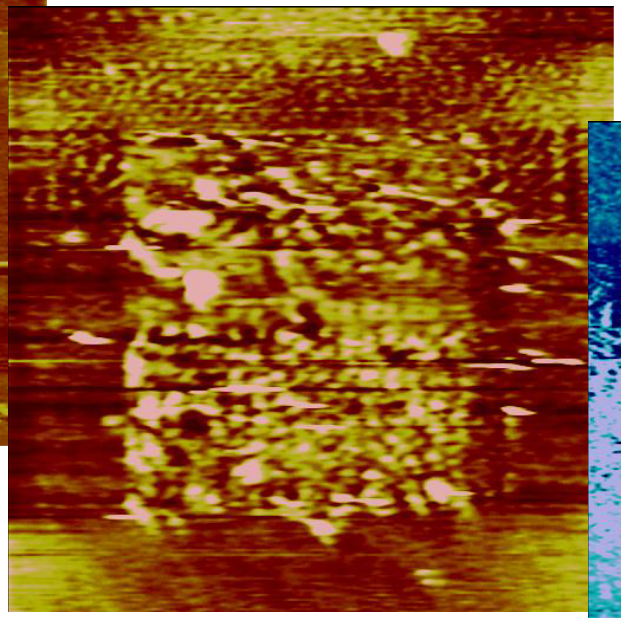




- Drift in contact resonance frequencies
- Training period (response maximizes after several cycles)

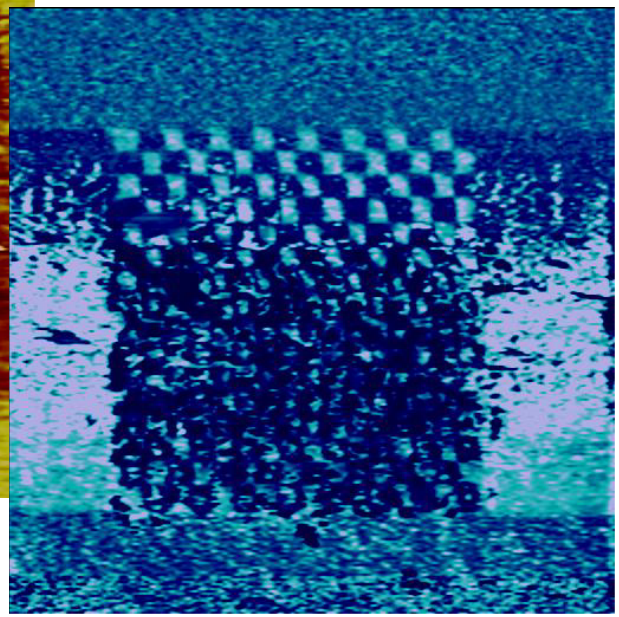
# Surface Damage in PFM



Surface topography after 5V writing at ~100% humidity



Surface topography after 10V writing at ~100% humidity



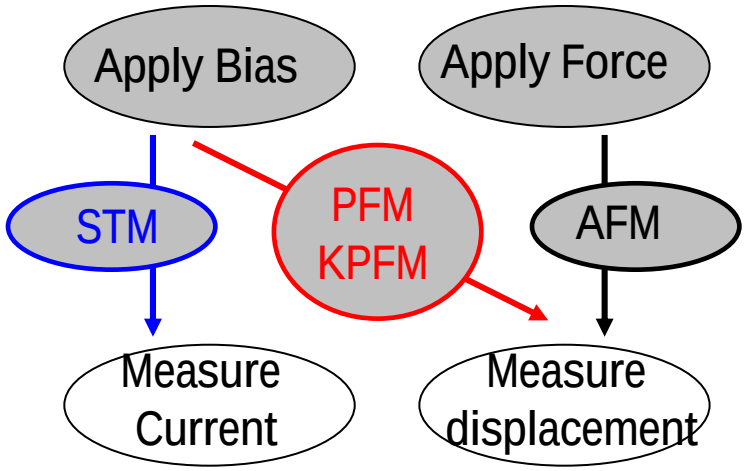
PFM image after 10V writing at ~100% humidity

Sometimes, even critical surface damage does not affect PFM!

# Energy Dissipation in SPM

## *Applications*

All energy transformation processes are associated with energy losses above thermodynamically necessary: local dissipation centers



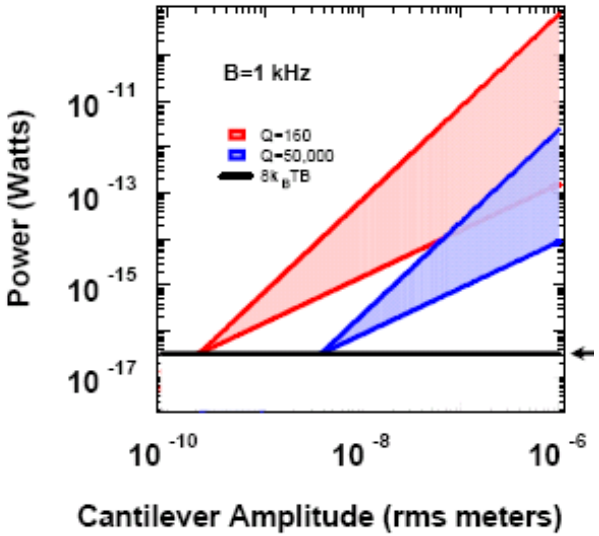

## *Fundamental Physics*

- Carrier-impurity scattering (e.g. Drude theory)
- Magnetoacoustic phonon generation
- Molecular vibrations
- Electron-phonon coupling
- Quasiparticles

Detection Limit (thermal vibration):

$$P^* = 8k_B T\Delta f \approx 3.3\cdot 10^{-17}\,\text{Watts}$$

or ~3 meV/oscillation

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