Dynamic behaviour in piezoresponse force microscopy

Stephen Jesse, Arthur P Baddorf and Sergei V Kalinin

Condensed Matter Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA
E-mail: sergei2@ornl.gov

Received 26 November 2005, in final form 3 January 2006
Published 21 February 2006
Online at stacks.iop.org/Nano/17/1615

Abstract

Frequency-dependent dynamic behaviour in piezoresponse force microscopy (PFM) implemented on a beam-deflection atomic force microscope (AFM) is analysed using a combination of modelling and experimental measurements. The PFM signal is comprised of contributions from local electrostatic forces acting on the tip, distributed forces acting on the cantilever, and three components of the electromechanical response vector. These interactions result in the flexural and torsional oscillations of the cantilever, detected as vertical and lateral PFM signals. The relative magnitudes of these contributions depend on geometric parameters of the system, on the stiffnesses and frictional forces of the tip–surface junction, and on the frequency of operation. The dynamic signal formation mechanism in PFM is analysed and conditions for optimal PFM imaging are formulated. An experimental approach for probing cantilever dynamics using frequency–bias spectroscopy and deconvolution of electromechanical and electrostatic contrast is implemented.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

In the last decade, piezoresponse force microscopy has become established as a powerful tool for probing local electromechanical activity on the nanometre scale [1–4]. Developed originally for imaging domain structures in ferroelectric materials, PFM was later extended to local hysteresis loop spectroscopy [5, 6] and ferroelectric domain patterning for applications such as high density data storage [7, 8] and ferroelectric lithography [9–11]. It was suggested recently that vector PFM can be used to determine local molecular or crystallographic orientation in piezoelectric materials, provided that all three components of the electromechanical response vector are determined quantitatively [12]. Broad applicability of PFM to materials such as ferroelectric perovskites [13, 14], piezoelectric III–V nitrides [15], and, recently, biological systems such as calcified and connective tissues [16–18], has necessitated fundamental theoretical studies of the image formation mechanism in PFM.

It was recognized that electrostatic tip–surface forces and buckling oscillations of the cantilever can provide significant and in some cases even dominating contributions to the PFM signal [19–21]. The imaging of ferroelectric materials in the vicinity of a phase transition at small probing biases or imaging of biological systems with weak electromechanical coupling require optimal imaging conditions to be established, and a number of approaches based on using contact resonances in PFM have been suggested [22, 23].

In our previous publications, we presented an in-depth analysis of the static (low frequency) PFM imaging mechanism and demonstrated approaches for data interpretation and visualization [12, 24, 25]. In particular, we have shown that under the condition of good tip–surface contact (no potential drop in the tip–surface gap), material properties measured by PFM are almost independent of the geometric characteristics of the tip, thus distinguishing this technique from mechanical SPM probes such as atomic force acoustic microscopy [26] or ultrasonic force microscopy [27, 28] in which contact stiffness scales linearly with tip–surface contact radius. However, to fully utilize PFM as a quantitative tool for local material

1 Author to whom any correspondence should be addressed.
characterization, an understanding of the dynamic behaviour, including the frequency-dependent contrast in vertical and lateral PFM, is required.

Here, we analyse the dynamic cantilever behaviour in PFM using both experimental 2D frequency–bias spectroscopy and elastic beam theory. We analyse the difference between signal transduction in the vertical PFM (VPFM) and lateral PFM (LPFM), discuss the contribution of longitudinal response to the VPFM signal, and discuss the contribution of cantilever buckling oscillations. Guidelines for quantitative PFM imaging and optimal frequency regimes are formulated.

2. Principles of PFM

Piezoresponse force microscopy is based on the detection of electrical bias-induced mechanical surface deformation [3, 4]. The conductive tip is brought into contact with the surface and a periodic bias, \( V_{\text{tip}} = V_0 + V_0 \cos(\omega t) \), is applied in which \( V_0 \) is the dc component of the tip bias and \( V_0 \) is the probing voltage. The bias results in a periodic surface displacement due to the piezoelectric effect. The components of the response vector at the modulation frequency, \( \omega \), are \( \{A, d_1, d_2, d_3\} \), corresponding to \( \{z, x, y\} \), or (vertical, longitudinal, lateral) directions of the coordinate system of the cantilever, as illustrated in figure 1. Note that the longitudinal component of surface displacement (in-plane component oriented along the longitudinal cantilever axis, \( d_2 \)) contributes to the vertical PFM signal [29]. In addition, bias generates local (acting on the tip) and non-local (acting on the cantilever) components of the electrostatic force resulting in flexural bending of the cantilever [19, 30]. These five contributions result in flexural and torsional oscillations of the cantilever beam and are detected by the optical system as vertical and lateral [31, 32] components of PFM signal. Note that the transduction of surface vibrations to the cantilever varies strongly for different components. While vertical surface vibrations are transferred to the tip, in-plane vibrations do not necessarily transfer due to the possible onset of sliding friction. Traditionally, the vertical PFM signal is measured in the units corresponding to purely vertical displacements of the tip normalized to driving voltage amplitude, i.e. the piezoresponse amplitude can be defined as \( A \equiv A_{\text{el}} + A_{\text{piezo}} + A_{\text{el}} \), where \( A_{\text{el}} \) is an electrostatic contribution due to tip–surface forces, \( A_{\text{piezo}} \) is an electromechanical contribution due to piezoelectric surface deformation and \( A_{\text{el}} \) is a non-local contribution due to capacitive cantilever–surface interactions, measured in units of \((\text{pm} \, V^{-1})\). Below, we address the relationship between the measured vertical signal, cantilever deflection angle, and surface displacement vector and local and distributed force components.

The contrast formation mechanism in PFM is determined by the interplay of contact mechanics of the tip–surface junction and cantilever dynamics. The mechanical equivalent circuit is a cantilever beam of length \( L \), with one end fixed, and the other end connected to lateral and vertical springs having spring constants \( k_1, k_2, \) and \( k_3 \), as shown in figure 1. This contact model has been broadly considered in the context of atomic force acoustic microscopy, and in the Hertzian approximation [33], the vertical spring constant, \( k_1 = 2aE^* \), where \( a \) is contact radius and \( E^* \) is the reduced Young’s modulus of the surface. The lateral spring constant of the tip–surface junction is \( k_2 = k_3 = 8aG^* \), where \( G^* \) is the reduced shear modulus. For isotropic materials, the ratio between the vertical and lateral spring constants is determined solely by the Poisson ratio, \( \nu \), and is given by \( k_1/k_3 = (2 - \nu)/(2 - 2\nu) = 1.2 \). For piezoelectric materials, the contact mechanics are voltage dependent and a rigorous solution is currently available only for transversely isotropic materials [24, 25].

3. Theory

3.1. Contributions to PFM contrast

A cantilever in combination with an optical beam deflection detector is the key element of an AFM force detection system. The motion of the cantilever induced by surface oscillations has been studied extensively in the context of atomic force acoustic microscopy (AFAM) [34–36] and ultrasonic force microscopy (UFM) [37]. However, electrostatic modulation in PFM gives rise to additional local and non-local force contributions and the boundary conditions are different. The analysis of the dynamic image formation mechanisms in PFM should include the following contributions as illustrated in figure 1:

1. the local vertical surface displacement \( (d_1) \);
2. the longitudinal, in-plane surface displacement along the cantilever axis \( (d_2) \);
3. lateral surface displacement, in-plane and perpendicular to the cantilever axis \( (d_3) \);
4. the local electrostatic force acting on the tip \( (f_{el}) \);
5. the distributed electrostatic force acting on the cantilever \( (q) \).

Below, we analyse the dynamic behaviour of the PFM probe. The cantilever is modelled as a uniform beam parallel to the surface. The solutions for more realistic scenarios, including cantilever tilt and damping, while easy to incorporate, are significantly more cumbersome, obviating insight into the imaging mechanism.
The general solution for cantilever dynamics in PFM is derived in section 3.2. In section 3.3, the important case of cantilever vibration under purely electrostatic forces is considered, providing an approach for probe calibration. Sections 3.4 and 3.5 discuss the limiting cases of zero and infinite lateral stiffness constants, corresponding to frictionless contact and cantilevered piezo-indentation. Finally, the torsional response of the cantilever is briefly analysed in section 3.6.

3.2. Cantilever dynamics in PFM

The dynamic behaviour of the cantilever is described by the beam equation

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

(1)

where \(u\) is the vertical displacement, \(x\) is the position along the cantilever, \(E\) is the Young’s modulus of the cantilever material, \(I\) is the second moment of inertia of the cross-section, \(\rho\) is the density, \(S_c\) is the cross-sectional area, and \(q(x,t)\) is the distributed force acting on the cantilever. For a rectangular cantilever, \(S_c = w \cdot h\) and \(I = wh^3/12\), where \(w\) is the cantilever width and \(h\) is the thickness. The cantilever spring constant, \(k\), is related to the geometric parameters of the cantilever by \(k = 3EI/L^3 = Ewh^3/4L^3\).

Note that in most commercial instruments the vertical deflection is measured in units of length (nm), even though the photodetectors measure the deflection angle. The relationship between the two is established using a suitable calibration procedure. For a purely vertical displacement, the relationship between cantilever deflection angle and measured height is \(u(L) = 2\theta(L)L/3\), relating the static vertical displacement of the cantilever end \(u(L)\) with the cantilever deflection angle \(\theta(L)\) [38]. However, in the dynamic case and for in-plane displacements, this relationship fails and the ratio between the deflection angle and tip displacement is now dependent on the type of the vibration mode and mode number. In order to relate the calculated dynamic signal to the amplitudes measured by AFM, the response amplitude \(A = 2\theta(L)L/3\) is defined, where the deflection angle of the cantilever is related to the local slope as \(\theta(L) = \arctan(u'(L)) \approx u'(L)\).

Equation (1) is solved in the frequency domain by introducing \(u(x,t) = u_0(x)e^{i\omega t}, q(x,t) = q_0e^{i\omega t}\), where \(u_0\) is the displacement amplitude, \(q_0\) is a uniform load per unit length, \(t\) is time, and \(\omega\) is modulation frequency. After substitution, equation (1) becomes

\[
\frac{d^4 u_0}{dx^4} + \frac{\rho S_c}{EI} \frac{d^2 u_0}{dt^2} = \frac{q_0}{EI},
\]

(2)

where \(\kappa = \omega^2 \rho S_c / EI\) is the wavevector. On the clamped end of the cantilever, the displacement and deflection angle are zero, yielding the boundary conditions

\[
u_0(0) = 0 \quad \text{(3a)}
\]

and

\[
u_0'(0) = 0. \quad \text{(3b)}
\]

On the supported end and in the limit of linear elastic contact, the boundary conditions for the moment and shear force are

\[
EIu_0''(L) = k_2H(d_2 - u_0(L)H) \quad \text{(4a)}
\]

and

\[
EIu_0''(L) = -f_0 + k_1(u_0(L) - d_1) \quad \text{(4b)}
\]

where \(f_0\) is the first harmonic of the local force acting on the tip and \(H\) is the tip height. For non-piezoelectric materials, \(d_1 = d_2 = 0\), while for zero electrostatic force, \(f_0 = 0\), providing purely electromechanical and electrostatic limiting cases for equation (1).

After solving the linear equation (2) and using \(EI = kL^3/3\), the deflection angle at the end of the beam \((x = L)\) is

\[
\theta(\beta) = \frac{A_\epsilon(\beta)q_0 + A_i(\beta)q_0}{N(\beta)} \quad \text{(5)}
\]

where

\[
A_\epsilon(\beta) = 3\beta^4 k_1 k L \sin \beta \sin \beta \quad \text{(6)}
\]

\[
A_i(\beta) = 3\beta^2 H k_2 (3k_1 + \cos \beta - 3k_1 \cos \beta + \beta^2 k \sin \beta) + \beta^2 k \cos \beta \sin \beta \quad \text{(7)}
\]

\[
A_s(\beta) = 3L^2 (3k_1 \cos \beta - \cos \beta - k \beta^3 \sin \beta + (k \beta^3 + 3k_1 \sin \beta) \sin \beta) \quad \text{(8)}
\]

\[
N(\beta) = \beta^2 (9H^2 k_1^2 + \beta^4 k^2 L^2 + \cos \beta ((-9H^2 k_1^2 + \beta^4 k^2 L^2) \cos \beta + 3\beta k (k_1 L^2 + k_1^3 L^2) \sin \beta + 3\beta k (-k_1 L^2 + k_1^3 L^2) \cos \beta \sin \beta) \quad \text{(9)}
\]

and the dimensionless wavenumber is \(\beta = kL\). The resonant frequencies are given by the roots, \(\beta_n\), \(n = 1, 2, \ldots, \) of \(N(\beta) = 0\). The corresponding eigenfrequencies are \(\omega_n^2 = EI\beta_n^2 / \rho S_c L^4 = EH^2 \beta_n^4 / 12 L^4\). In terms of the cantilever spring constant, the resonant frequencies are \(\omega_n^2 = k\beta_n^2 / 3m\), where \(m = hLw\rho\) is the cantilever mass.

The terms \(A_\epsilon(\beta)/N(\beta), A_i(\beta)/N(\beta), A_s(\beta)/N(\beta)\) describe the frequency dependence of the PFM signal due to vertical and longitudinal components of the surface displacement, the local electrostatic force acting on the tip, and the distributed electrostatic force acting along the cantilever, respectively. The respective PFM signal components are \(A_{el} = 2A_\epsilon(\beta)f_0 L/3N(\beta), A_{piezo} = 2(A_\epsilon(\beta)d_1 + A_i(\beta)d_2)f_0 L/3N(\beta),\) and \(A_{al} = 2A_s(\beta)q_0 L/3N(\beta)\). Note that the vertical electromechanical contribution and local force contribution have similar frequency dependences (compare equations (6) and (8)).

The resonant frequencies of the system described by equation (5) are determined only by the (voltage-dependent) mechanical properties of the system and are independent of the relative contributions of the electrostatic and electromechanical interactions. Therefore, tracking the resonant frequency of an electrically excited cantilever provides information on the local elastic properties of the sample, which is similar to frequency detection in AFAM, as analysed in detail by Rabe [39]. Moreover, in a linear model, vertical, longitudinal, and local electrostatic responses to the PFM signal cannot be separated by a proper choice of driving frequency. Therefore,
unambiguous measurement of all three components of the electromechanical response vector requires an alternative approach, for example based on either 3D SPM [40] or sample rotation [31].

In the low frequency limit, \( \beta \to 0 \), these contributions become

\[
A_e = \frac{6k_1 k L}{4k(k + k_1)L^2 + 3H^2 k_2(4k + k_1)} \approx \frac{6k L}{4kL^2 + 3H^2 k_2} \quad (11)
\]

\[
A_f = \frac{3H k_2(k + k_1)}{4(k + k_1)L^2 + 3H^2 k_2(4k + k_1)} \approx \frac{3H k_2}{4kL^2 + 3H^2 k_2} \quad (12)
\]

\[
A_d = \frac{6k L}{4k(k + k_1)L^2 + 3H^2 k_2(4k + k_1)} \approx \frac{k_1(4k L^2 + 3H^2 k_2)}{16(k + k_1)L^2 + 12H^2 k_2(4k + k_1)} \approx \frac{L^2}{4(4kL^2 + 3H^2 k_2)} \quad (13)
\]

Equations (11)–(14) thus describe the static deflection at frequencies well below the first resonance. Note that the relative magnitudes of these contributions are, as expected, sensitive to the length of the cantilever and, for stiff cantilevers where \( k L^2 \gg H^2 k_2 \), the response is determined primarily by the vertical displacement of the tip. For a soft cantilever, the in-plane displacement of the tip apex is the dominant oscillation mode. From equations (11)–(14), the scaling of the electromechanical and electrostatic responses with the geometric parameters of the cantilever (\( L, H \)) and with the spring constants \((k, k_1, k_2)\) can be determined in a straightforward manner.

According to equations (5)–(10), the frequency dependence of individual contributions is

\[
A_e(\beta) = \frac{k_1}{k^2} \frac{1}{\omega} \quad (15)
\]

\[
A_f(\beta) = \frac{1}{\kappa} \approx \frac{1}{\omega^{3/2}} \quad (16)
\]

\[
A_d(\beta) = \frac{k_1}{k^2} \frac{1}{\omega} \quad (17)
\]

\[
A_d(\beta) = \frac{1}{\kappa} \approx \frac{1}{\omega^{3/2}} \quad (18)
\]

From equations (15)–(18), all four contributions decrease with frequency due to the dynamic stiffening effects. Even in the absence of damping, the non-local contribution scales as a higher power of frequency, suggesting that non-local cantilever effects will be minimized at high frequencies. At the same time, the local electrostatic and normal electromechanical contributions scale in a similar manner as the ratio, \( A_{piezo}/A_d = \frac{d_1 k_1}{f_0} \), which depends only on the spring constant of the tip–surface junction. This suggests that these contributions cannot be distinguished by a choice of the operating frequency. Instead, either the use of a cantilever with a high spring constant at high indentation forces \((k_1 \to \infty)\), imaging at the nulling bias, or using shielded probes [41] (where \( f_0 \to 0 \)) is required. In the linear elastic approximation, the dynamic stiffening effect is less pronounced for the longitudinal signal; however, the onset of sliding friction can minimize signal transduction. This behaviour is analysed in more detail in section 4.2.

The general frequency dynamics of PFM, as described by equation (5), are complex and depend on the vertical and longitudinal spring constants of the tip–surface junction and geometric parameters of the cantilever. However, a number of important limiting cases can be analysed, as summarized below.

3.3. Cantilever dynamics under electrostatic forces

One important limiting case is that of the flexural oscillations of a supported cantilever due to a distributed electrostatic force (buckling oscillations). In this case, \( k_1 \to \infty \) and \( k_2 \to \infty \) in equation (5), and the boundary conditions on the supported end are

\[
u_0(L) = 0 \quad (19a)
\]

and

\[
u''_0(L) = 0, \quad (19b)
\]

corresponding to zero displacement and no applied rotational moment. The frequency response of such a supported cantilever is

\[
\theta(\beta) = \frac{3q_0 \cos \beta - \cosh \beta + \sin \beta \sinh \beta}{k \beta^2} \cos \beta \sin \beta - \cos \beta \sinh \beta \quad (20)
\]

The lowest order resonances in equation (20) correspond to \( \beta_n = 3.927, 7.067, 10.21 \). In the low frequency limit \( (\beta \to 0) \), \( \theta = q_0/16k \) and the PFM signal is \( A = q_0 L/24k \).

For comparison, for a free cantilever (tip not touching the surface) that is under the action of distributed and local forces, the boundary conditions on the free end are

\[
u''_0(L) = 0, \quad (21a)
\]

and

\[
E 1 \nu''_0(L) = -f_0. \quad (21b)
\]

The cantilever dynamic response in this case is

\[
\theta(\beta) = -\frac{3Lq_0 \sin \beta + 3(Lq_0 + \beta f_0 \sin \beta) \sinh \beta}{k^3 \beta (1 + \cosh \beta \cos \beta)} \quad (22)
\]

At low frequencies \( (\beta \to 0) \), the quasi-static cantilever deflection is \( \theta = (q_0/2 + 3f_0/2L) \), and the measured PFM signal is \( A = (q_0 L/3k + f_0/k) \). For a free cantilever, the response due to the non-local force is eight times larger than that for the clamped cantilever. The resonances in equation (22) occurs for \( \beta_n = 1.875, 4.694, 7.855 \).

Given that the resonant frequencies for the electrostatically driven free cantilever are independent of the ratio between the distributed and localized forces, electrostatic excitation can be used for cantilever calibration in PFM measurements, providing a complementary approach to cantilever calibration using mechanically induced resonances [42–44].
3.4. Frictionless contact

An important limiting case for probe dynamics in PFM corresponds to frictionless contact between the tip and the surface, \( k_2 \to 0 \), also equivalent to \( u''_0(L) = 0 \) in equation (4a). In this limit, the response is

\[
A_v(\beta) = 3\beta^4k_1k\sin\beta\sinh\beta \quad (23a)
\]

\[
A_i(\beta) = 0 \quad (23b)
\]

\[
A_q(\beta) = 3L(3k_1(\cos\beta - \cosh\beta) - k\beta^3\sin\beta + (k\beta^3 + 3k_1\sin\beta)\sinh\beta) \quad (23c)
\]

\[
A_{\theta q}(\beta) = \beta^3kL(\beta^3k + \cosh\beta(3\beta^3k\cos\beta + 3k_1\sin\beta) - 3k_1\cos\beta\sinh\beta) \quad (23d)
\]

If, additionally, the vertical spring constant of the tip–surface junction is large compared to that of the cantilever, \( k_1 \to \infty \), equations (23a)–(23e) simplify to

\[
\theta(\beta) = \frac{3Lq_0(\cos\beta - \cosh\beta) + (d_1k\beta^4 + Lq_0)\sin\beta\sinh\beta}{\beta^3kL(\cos\beta\sin\beta - \cosh\beta\sin\beta)} \quad (24)
\]

Equation (24) describes the cantilever dynamics for the case of a soft cantilever on a piezoelectric surface under the effect of both a piezoelectric deformation and a distributed load.

In a more realistic case, when the cantilever can slide along the surface, the frictional force will be non-zero. An analysis of this linear force is reported elsewhere \([45]\). A rigorous description of this process for a periodic surface motion yields a non-linear equation of motion and requires quantitative models of lateral contact mechanics of the tip surface junction \([46–50]\). Here, these effects will be addressed experimentally in section 5. Also note that the inclination of the surface with respect to cantilever will lead to coupling between normal and lateral contributions, for example as analysed by Rabe for AFAM \([39, 51]\).

3.5. The stiff cantilever limit

In the case when the distributed force is ignored and only the vertical displacement at the tip–surface junction is considered (as is generally the case with a stiff cantilever, \( kL^2 \gg H^2k_2 \)), the angle of the tip is given by

\[
\theta(\beta) = [3\beta(f_0 + d_1k_1)\sin\beta\sinh\beta][L(\beta^3k + \cosh\beta(\beta^3k\cos\beta + 3k_1\sin\beta) - 3k_1\cos\beta\sinh\beta)]^{-1} \quad (25)
\]

Equation (25) thus describes the frequency dynamics in the most common case of a PFM measurement performed with a stiff cantilever and with a beam-deflection detector. In the quasistatic regime, equation (25) becomes \( \theta = \frac{3(f_0 + d_1k_1)/2k_1L}{3d_1/2L} \), i.e., a purely electromechanical response. For a free cantilever, \( k_1 \to 0 \), the response is \( \theta = -3f_0/2kL \) and \( A = -f_0/k_1 \), i.e., a purely electrostatic response. In the contact regime for a non-piezoelectric material, \( \theta = -3f_0/2k_1L \) and the measured deflection is \( A = -f_0/k_1 \), i.e., the measured displacement scales reciprocally with the tip–surface junction spring constant, \( k_1 \), and, as expected, is independent of the cantilever length, \( L \). For non-zero frequencies in the limit \( k_1 \to \infty \), equation (25) becomes

\[
\theta(\beta) = \frac{-d_1\beta\sin\beta\sinh\beta}{L(\cosh\beta\sin\beta - \cos\beta\sinh\beta)} \quad (26)
\]

whereas for \( k_1 \to 0 \), equation (25) becomes (5) for a free cantilever. Equation (26) thus describes the contact dynamics of PFM in the purely electromechanical case.

Note that the resonant frequencies for the cantilever (figure 2(b)) increase with the spring constant of the tip–surface junction, indicative of the stiffening of the corresponding mode. The individual branches are separated by frequency gaps, as has been shown by Rabe \([34]\). The cross-over from free cantilever modes to supported cantilever modes shifts to high contact spring constants with the resonance number (dotted line in figure 2(b)), as shown by Turner \([52]\). The corresponding diagram for the longitudinal response is shown in figure 2(c), demonstrating similar behaviour. Thus, imaging at higher resonant frequencies enhances the sensitivity to the elastic properties of materials with high elastic moduli.

3.6. Lateral contribution

In lateral PFM, torsional oscillations of the cantilever are detected. The distributed and localized electrostatic forces are generally directed along the surface normal and are symmetric with respect to the longitudinal axis of the cantilever and therefore provide minimal contribution to the torsional behaviour. Thus, the torsional oscillations can be considered to be primarily induced by surface displacement. The torsional oscillations have been extensively studied in the context of frictional acoustic force microscopy \([53, 54]\). Here, we briefly repeat this analysis for PFM.

The equation of motion for torsional oscillation is

\[
c_T\frac{\partial^2\Theta}{\partial t^2} = \rho J\frac{\partial^2\Theta}{\partial \xi^2} = 0 \quad (27)
\]

where \( \Theta \) is the angle of torsion, \( J \) is the second moment of inertia and \( c_T \) is the torsional stiffness. For a rectangular beam, \( c_T = wh^3G/3 \), where \( G \) is the shear modulus of cantilever material. In the frequency domain, equation (27) becomes

\[
\frac{\partial^2\Theta(\xi)}{\partial \xi^2} + \xi^2\Theta(0) = 0 \quad (28)
\]

where \( \xi^2 = \rho J\omega^2/c_T \) and \( \Theta(x, \xi) = \Theta_0(x)e^{i\omega\xi} \). The boundary condition on the clamped end is \( \Theta_0(0) = 0 \). On the supported end, the boundary conditions are obtained in a similar way as with longitudinal oscillations. For the case of a linear elastic contact, the boundary condition on the supported end is

\[
\Theta_0(L) = -\tilde{k}_3(H\Theta_0 - d_3) \quad (29)
\]

where \( d_3 \) is the in-plane displacement of the surface perpendicular to the cantilever long axis and \( \tilde{k}_3 = Hk_3/c_T \). The solution in this case becomes

\[
\Theta_0(L) = \frac{d_3\tilde{k}_3\sin\xi L}{\xi\cos\xi L + H\tilde{k}_3\sin\xi L} \quad (30)
\]

For an infinitely stiff contact, \( k \to \infty \) and \( \Theta_0(L) = d_3/H \), while for a free cantilever, \( k_3 \to 0 \) and \( \Theta_0(L) = 0 \), as expected. Note that the resonant frequencies, as determined by the denominator of equation (30), depend on the stiffness constant, \( k_3 \) (figure 2(d)).
4. Implications

In this section, we analyse the implications of frequency-dependent probe dynamics on PFM. In section 4.1, the electrostatic cantilever contribution to the measured signal is analysed. The frequency-dependent dynamics of the cantilever are analysed in section 4.2. Finally, section 4.3 discusses the relationship between the phase shift between domains and the relative magnitude of the electrostatic contribution.

4.1. Cantilever contribution

The cantilever–surface capacitive force in the plane–plane model can be approximated as $F_{\text{cap}} = \varepsilon_0 S (V_{\text{tip}} - V_{\text{surf}})^2 / 2H^2$, where $S = Lw$ is the cantilever area [55]. The first harmonic of the distributed force is thus $q_0 = \varepsilon_0 S V_{\text{ac}} \Delta V / 2LH^2$, where $H$ is the tip height (equal to the cantilever–surface separation) and $\Delta V = V_{\text{dc}} - V_{\text{surf}}$. The non-local contribution to the PFM signal is $A_{\text{nl}} = -Lw\varepsilon_0 V_{\text{ac}} \Delta V / 48kH^2$, and the overall PFM signal in the low frequency regime is

$$A = d_1 + \frac{Lw\varepsilon_0 V_{\text{ac}} \Delta V}{48kH^2}, \quad (31)$$

where the first term is the electromechanical contribution, and the second term is the non-local contribution due to the cantilever buckling oscillations. Note that the non-local contribution is inversely proportional to the cantilever spring constant, while the electromechanical contribution is spring constant independent.

As discussed above, PFM imaging and quantitative piezoresponse spectroscopy require the electromechanical interaction to be much stronger than that of the non-local electrostatic interaction. From equation (31), this condition can be derived as $k \gg Lw\varepsilon_0 \Delta V / 48d_1H^2$. By taking the estimates, $d_1 = 50 \text{ pm V}^{-1}$, $\Delta V = 5 \text{ V}$, $L = 225 \mu m$, $w = 30 \mu m$, $H = 15 \mu m$, the lower limit of the spring constant is given by $k > 0.55 \text{ N m}^{-1}$. Note that while for a zero tip–surface potential difference, $\Delta V = 0$, the non-local interactions are formally absent, this condition is rarely achieved experimentally, unless a top-electrode experimental set-up is used [3]. This electrostatic contribution also precludes the determination of electrostrictive constants from PFM hysteresis measurements because both electrostrictive and electrostatic contributions scale linearly with $\Delta V$ and thus cannot be distinguished unambiguously.

4.2. Frequency regimes in PFM

Here, we analyse the frequency dependence of the dominant contrast mechanism in the vertical PFM signal using the result in section 2. The response is calculated for three cantilevers with $L = 224 \mu m$, $w = 24 \mu m$, $H = 15 \mu m$, and thicknesses of $h = 1.05, 3$, and $8 \mu m$, corresponding to spring constants of 0.1 N m$^{-1}$ (cantilever A), 2.4 N m$^{-1}$ (cantilever B) and 40 N m$^{-1}$ (cantilever C), as summarized in table 1. The piezoelectric constant of the sample is taken to be $d_1 = 5 V_{\text{ac}}$ (pm), corresponding to a weakly piezoelectric material. The local electrostatic force is taken to
be 0.7 nN V\(^{-1}\). The spring constant of the tip–surface junction in all cases was 1000 N m\(^{-1}\) (load \(\approx\) 100 nN) and assumed to be independent of the cantilever spring constant. These conditions correspond to the case when the indentation force is dominated by capillary and adhesive forces, typical for imaging under ambient conditions (note that using constant deflection will result in a qualitatively different behaviour, when the spring constant of the tip–surface junction depends on the cantilever spring constant).

Figures 3(a), (c) and (e) (left) show the PFM amplitude, \(A_{\text{piezo}} + A_{\text{elec}} = 2L_0 \theta(\beta)/3\), maps as a function of frequency and tip–surface potential difference, \(\Delta V\), calculated according to equation (26) for a zero local electrostatic force, \(f_0 = 0\). A number of resonances (bright lines) and anti-resonances (black lines) can be clearly seen. The phase changes by 180\(^\circ\) across resonance and anti-resonance lines. For low tip biases, the response is purely electromechanical and is independent of \(\Delta V\). For higher dc biases, the response is dominated by non-local contributions and is linear in \(\Delta V\). Note that the position of the resonances is determined solely by the cantilever properties and spring constant of the tip–surface junction and is independent of the tip bias. At the same time, the anti-resonances on the response diagram are strongly bias dependent. Noteworthy is the fact that the nulling bias, \(V_{\text{null}}\), corresponding to the zero of the response, provides a measure of the electrostatic contribution to the signal. Indeed, in the electromechanical limit, the response is bias independent, and the nulling bias is infinite, while in the purely electrostatic limit, the nulling bias is equal to the surface potential, \(V_{\text{surf}}\). In the intermediate case, the nulling bias is \(V_{\text{null}} = V_{\text{surf}} + a(\omega)d_1\), where \(a(\omega)\) is the frequency-dependent proportionality constant, \(a(\omega) = A_0/A_0\). Therefore, the frequencies for which \(V_{\text{null}}\) is maximal correspond to the frequencies at which the electromechanical contrast is dominant, and the resulting PFM image has optimal contrast.

The relative magnitudes of the non-local and electromechanical contributions, \(A_{\text{piezo}}/(A_{\text{piezo}} + A_{\text{elec}})\), are illustrated in figures 3(a), (c) and (e) (right). The white region corresponds to a dominant electromechanical contrast, while black regions correspond to dominant non-local electrostatic contributions. In the low frequency limit, the crossover between the two (indicated by an arrow) scales proportionally to the cantilever spring constant, as follows from the analysis in section 4.1. At high frequencies, the relative contribution of the electromechanical contrast increases, indicative of dynamic cantilever stiffening. Note that the resonances do not affect the relative contributions of these signals, thus justifying the applicability of contact resonance-enhanced PFM imaging to low coercive bias materials.

In the presence of a non-zero local electrostatic force, the response diagrams for PFM signal including local, non-local, and electromechanical contributions, \(A = A_{\text{piezo}} + A_{\text{elec}} + A_{\text{local}}\), can be constructed as shown in figures 3(b), (d) and (f). The intensity of red, green, and blue components represents the relative contributions of local electrostatic (LE), non-local electrostatic (NL), and electromechanical (PE) components, defined as, for example, \(A_{\text{piezo}}/(A_{\text{piezo}} + A_{\text{elec}} + A_{\text{local}})\) for the electromechanical component. The position of the boundary between the local electrostatic and electromechanical contributions is frequency independent, as expected.

While these results are specific to the chosen parameters, similar diagrams can be readily constructed for different geometric parameters of the system and material properties. However, in all cases, the qualitative features of figure 3 are valid, including the preponderance of the electromechanical response at low biases, the dominance of the non-local contribution in the vicinity of anti-resonances, the minimization of the non-local cantilever contribution at high frequencies, and the frequency-independent ratio between local electrostatic and electromechanical contributions determined only by the spring constant of the tip–surface junction.

### Table 1. Cantilever properties.

| Cantilever | \(w\) (\(\mu\)m) | \(h\) (\(\mu\)m) | \(L\) (\(\mu\)m) | \(k\) (N m\(^{-1}\)) |
|------------|-----------------|-----------------|-----------------|-----------------|
| A          | 24              | 8               | 224             | 45              |
| B          | 24              | 3               | 224             | 2.4             |
| C          | 24              | 1.05            | 224             | 0.1             |

4.3. Phase shifts between anti-parallel domains

One of the suggested guidelines for quantitative PFM imaging is the requirement for the phase shift between opposite domains to be 180\(^\circ\), with equal amplitudes on both sides of the domain wall [19]. In the low frequency case this generally holds true; the phase shift between domains is always 180\(^\circ\) or 0\(^\circ\), depending on the relative magnitudes of electrostatic and electromechanical contributions. In the dynamic case however, the electromechanical and electrostatic contributions to the PFM signal have different phases. Defining the PFM x-signal as \(PR = A \cos \psi\), the response over \(c^+\) and \(c^-\) domains can be written as

\[
P R_{\pm} = \pm d_1 + G(V_{dc} - V_{surf}) \exp(i\psi)
\]  

where \(\psi\) is the phase difference between the electrostatic and electromechanical responses and \(G\) is the proportionality factor containing local and non-local electrostatic contributions to the signal, \(G = (A_0(\beta)C_{\text{tip}} + A_0(\beta)C_{\text{surf}})/N(\beta)\), where \(C_{\text{tip}}\) and \(C_{\text{surf}}\) are tip–surface and cantilever–surface capacitance gradients.

From equation (32), a small electrostatic force contribution, \(G(V_{dc} - V_{surf}) \rightarrow 0\), gives PFM amplitudes of

\[
P R_{\pm} = d_1 \pm G(V_{dc} - V_{surf}) \cos \psi
\]

and the phase difference is

\[
\Delta \psi = \pi - \sin(2\psi)(G(V_{dc} - V_{surf}))^2/d_1^2.
\]

\[33\]

Thus, the deviation of the phase shift between domains from 180\(^\circ\) provides a measure of the electrostatic contribution, and validates its use as a criterion for quantitative PFM.

In the limit of a small piezoelectric contribution, \(d_1 \rightarrow 0\), the domain contrast and phase change between the domains are respectively \(PR_{\pm} = G(V_{dc} - V_{surf}) \pm d_1 \cos \psi\) and

\[
\Delta \psi = 2 \sin(2\psi)d_1/(G(V_{dc} - V_{surf}))(\cos 2\psi - 3).
\]

\[34\]

From equations (33) and (34) the following picture emerges. In the purely electromechanical case, \(G_{\text{elec}}\) is identically zero. The response amplitudes are equal in the \(c^+\) and \(c^-\) domain regions, while the phase changes by 180\(^\circ\) between the domains. For domains with an arbitrary orientation, the absolute value of the amplitude signal provides a measure of the piezoelectric activity of the domain, and
5. Experiment

PFM was implemented on a commercial SPM system (Veeco MultiMode NS-III) equipped with additional function generators and lock-in amplifiers (DS 345 and SRS 830, Stanford Research Instruments, and Model 7280, Signal Recovery, frequency range 0–2 MHz). A custom-built sample holder was used to allow direct tip biasing and to avoid capacitive cross-talk with the SPM electronics. This holder does not contain a piezoactuator, thus reducing problems due to stray resonances. In most cases, the tip mount was glued to the holder using conductive epoxy to provide a rigid, electrically conductive connection. The detailed description of PFM and its experimental aspects is available elsewhere [2–4].

A bias (both dc and ac) was applied directly to the tip either in contact mode (producing both an electrostatic and an electromechanical contribution) or at a small distance above the surface (yielding a purely electrostatic contribution). Measurements were performed on non-piezoelectric SiO₂, periodically poled LiNbO₃, and polycrystalline lead zirconate titanate (PZT) ceramics using Pt coated and Au coated tips (NCSC-12 C, Micromasch, l ≈ 130 μm, resonant frequency ∼150 kHz, spring constant k ∼ 4.5 N m⁻¹), Co–Cr coated tips (Veeco, resonant frequency ∼72 kHz, spring constant k ∼ 1 N m⁻¹) and calibrated Nanosensors (Nanosensors GmbH) Pt coated tips.

To address PFM dynamics, the vertical and lateral cantilever responses were measured as a function of both tip dc bias and frequency, producing a 2D frequency–bias spectroscopic map (similar to the presentation of modelling results shown in figure 3).

6. Results and discussion

Experimental results of the force–bias spectroscopy of cantilever dynamics are presented in section 6.1. The
frequency dependence of the lateral PFM response is discussed in section 6.2. The bias effect on PFM imaging data is studied in section 6.3. An approach for deconvolution of electrostatic and electromechanical components is discussed in section 6.4.

6.1. PFM dynamics in contact and non-contact modes

Figure 4 illustrates the response acquired with a Co–Cr tip (tip 1) on a SiO2 and LiNbO3 surface in contact and non-contact modes. The resonances for SiO2 in the contact regime are \( \omega_1 = 407.9 \pm 0.3 \text{ kHz} \), and \( \omega_2 = 1075.02 \pm 0.15 \text{ kHz} \). The ratio of the resonant frequencies is \( \omega_2/\omega_1 = 2.63 \), lower than the theoretical ratio of 3.24:1. In the non-contact regime, the resonances are \( \omega_1 = 63.2 \pm 0.4 \text{ kHz} \), \( \omega_2 = 368.3 \pm 0.3 \text{ kHz} \), and \( \omega_3 = 1031.7 \pm 0.2 \text{ kHz} \). This gives the ratio of the first three harmonics as \( \omega_1: \omega_2: \omega_3 = 16.32:5.83:1 \), which is close to the theoretical ratio 17.55:6.23:1. Some discrepancy could be expected since, in the contact mode, the tip–surface interaction and absolute position of the tip along the cantilever significantly affect the resonance spectrum. In the non-contact regime, the behaviour is closer to the ideal model. Note that there are no zeros in the bias interval of study and the phase is constant along the bias axis, suggesting that the surface is strongly charged (\( V_{\text{surf}} > 8 \text{ V} \)). In the contact regime, the resonant frequencies are shifted compared to SiO2 (by 8.2 kHz for \( \omega_1 \) and 4 kHz for \( \omega_2 \)) due to the difference in elastic properties of the materials and the presence of an electromechanical response.

The vertical and lateral amplitude and phase response diagrams for SiO2 in non-contact (a), (b)) and contact (c), (d)) modes acquired using a Nanosensors tip (tip 2) \( (h = 3 \text{ m} \mu \text{m}, w = 24 \text{ m} \mu \text{m}, L = 223 \text{ m} \mu \text{m}, k = 2.4 \text{ N m}^{-1} \), as supplied by manufacturer) are shown in figure 5. The general structure of the non-contact resonances is similar to that in figure 4, with the major resonances at \( \omega_1 = 54.55 \pm 0.04 \text{ kHz} \), \( \omega_2 = 346.73 \pm 0.05 \text{ kHz} \), and \( \omega_3 = 980.53 \pm 0.1 \text{ kHz} \). The ratio of the frequencies is \( \omega_3/\omega_2/\omega_1 = 17.97:6.35:1 \), very close to the theoretical ratio 17.55:6.23:1. The frequency-independent nulling bias is \( V_{\text{tip}} = 1.0 \text{ V} \). The lateral response in figures 5(c) and (d) shows strong resonances at \( \omega_1 = 54.64 \pm 0.2 \text{ kHz} \), with a zero corresponding to \( V_{\text{tip}} = 1.0 \text{ V} \). Close similarities are evident between resonances in the vertical and lateral modes, suggesting that the latter is a result of the cross-talk between the normal and torsional cantilever oscillations. At higher frequencies, the lateral signal increases monotonically, presumably due to a cross-talk in the cabling. Response diagrams for the contact regime are shown in figures 5(e)–(h). The main resonances are similar to that for tip 1; however, an additional resonance at \( \omega = 634.1 \text{ kHz} \) emerges. The fact that the resonance frequency is sample independent and a nulling bias exists suggests that this resonance is due to the flexural mode in which the beam deflects in the plane parallel to the surface.

A similar response diagram measured for PZT in figures 5(i)–(l) shows a completely different behaviour. The nulling bias is now strongly frequency dependent, as expected for the case when the relative contributions of electrostatic and electromechanical signals vary due to their different frequency dependences (compare to figure 3).

This behaviour is further illustrated in figure 6. The resonant frequencies in the non-contact mode for SiO2 and LiNbO3 are identical, as illustrated in figure 6(a). In the contact regime, the resonant frequencies are shifted relative to SiO2 (by 8.2 kHz for \( \omega_1 \) and 4 kHz for \( \omega_2 \)) due to the difference in elastic properties of the materials (figure 6(b)). The frequency dependence of the vertical and lateral PFM signals in non-contact and contact regimes for SiO2 and PZT are summarized in figures 6(e) and (d). The frequency dependence of the vertical response is drastically different, due to a difference in the local elastic moduli and piezoelectric contributions to the
signal. At the same time, the lateral PFM signal, expected to be zero for SiO$_2$ and non-zero for PZT, is virtually identical at high frequencies and differences are observed only at low frequencies, suggesting a large cross-talk contribution at high frequencies.

6.2. Lateral PFM

The experimental data in figures 5, 6 illustrate that the frequency behaviour of the lateral signal differs strongly from that of the vertical. While in the latter case a number of cantilever resonances are observed in the frequency range of study (0–2 MHz), the frequency dependence of the lateral signal is relatively featureless (figures 5(c), (d) and 6(c), (d)). At high frequencies, the response is dominated primarily by capacitive cross-talk in the detector electronics and cabling. The differences in the lateral response of different PZT grains and SiO$_2$ are observed only at frequencies $\ll$100 kHz. The intrinsic lateral PFM image contrast was found to decay quickly at frequencies above $\sim$10 kHz (not shown), in agreement with results by Kholkin [56]. These results suggest that, for lateral PFM, effective detection is possible only at low (<10 kHz) frequencies. This response decays rapidly as the frequency increases, presumably due to the onset of sliding friction between the tip and the surface. These results also suggest that the longitudinal contribution to PFM contrast will be absent at high frequencies, due to similarities in the vibration transduction mechanisms for longitudinal and lateral components of in-plane surface displacement.

6.3. Bias effect on PFM imaging

To analyse the effect of bias conditions on the PFM signal, figure 7 illustrates the amplitude and phase images of a polycrystalline PZT surface at biases of 0, $-8$, and 8 V. Note that, for the image close to the nulling potential, the phase changes by 180° between domains. For the case of a strong electrostatic contribution, the phase is purely positive or negative, as expected. The relative response amplitudes in grains 1 and 2 change with tip bias, in agreement with the analysis in section 4.3.

Further insight into the frequency-dependent dynamics of PFM can be obtained by comparing the amplitude–frequency–bias response diagrams acquired from regions with dissimilar domain orientations (figure 8). Note that the orientation of the line corresponding to the frequency dependence of the

![Figure 5. Frequency–bias amplitude ((a), (c), (e), (i), (k)) and phase ((b), (d), (f), (h), (j), (l)) response diagrams for SiO$_2$ in the non-contact mode ((a)–(d)), SiO$_2$ in the contact mode ((e)–(h)), and PZT in the contact mode ((j)–(l)). Vertical ((a), (b), (e), (f), (i), (j)) and lateral ((c), (d), (g), (h), (k), (l)) response diagrams are shown.](image-url)
nulling bias (vertical dark line) is opposite for these two grains, indicative of the opposite signs of the electromechanical contribution to the PFM signal of anti-parallel grains.

The effect of tip wear on the frequency–bias spectrum is illustrated in figures 8(c) and (d), acquired after several repetitive topographic scans. Note that $V_{\text{null}}$ is strongly bias dependent for a good tip (figure 8(a)). For a deteriorated tip, the electromechanical contribution to the signal is small, so that $V_{\text{null}} \approx V_{\text{surf}}$, and is virtually frequency independent.

6.4. Deconvolution of the electrostatic and electromechanical contributions
The amplitude and phase response diagrams allow the electromechanical and electrostatic contributions to the PFM
signal to be distinguished. Briefly, the PFM $x$-signal, defined as $PR = A \cos \varphi$, can be represented as

$$PR_x = \tilde{d}_1 + \tilde{G}_{\text{elec}}(V_{dc} - V_{surf}),$$  \hspace{1cm} (35)

where $\tilde{d}_1$ and $\tilde{G}_{\text{elec}}$ are the electromechanical and electrostatic contributions now including a frequency-dependent phase multiplier. From equation (35), the electrostatic contribution to the PFM signal can be determined from the slope, $\tilde{G}_{\text{elec}} = c$, of the response versus bias curve at each frequency. The electromechanical contribution is related to the intercept, $b$, as $\tilde{d}_{\text{eff}} = b + cV_{surf}$. While the electrostatic contribution can be determined unambiguously, the electromechanical contribution depends on a known surface potential, $V_{surf}$, which can be determined for example from non-contact measurements.

Figure 9 shows the bias- and frequency-dependent response diagrams for non-piezoelectric SiO$_2$ in non-contact (a) and contact (c) regimes, as compared to PZT grain 2 acquired with new (e) and deteriorated (g) tips. The function $y = b + cV_{tip}$ was fitted to the signal for each frequency. Shown in figures 9(b), (d), (f) and (h) are error maps defined as $PR - (b + cV_{tip})$, representing the deviation of the actual response from a purely linear response. The scale for figures 9(b), (d), (f) and (h) is $1\%$ of full scale for figures 9(a), (c), (e) and (g). Note that the deviation from linearity is extremely small, suggesting the validity of equation (35). The maximum deviations are observed in the vicinity of resonances, where non-linear effects become pronounced.

The frequency dependences of the electromechanical and electrostatic responses for these materials are shown in figures 10(a) and (b). Note that the electromechanical response is greatest for PZT grain 1 with a good tip, slightly smaller for grain 2, and is negligibly small for SiO$_2$ in the non-contact and contact modes, as expected. In comparison, the electrostatic response is comparable for all materials (figure 10(b)). An alternative approach [23] based on measurements of an amplitude–frequency curve of piezoelectric and non-piezoelectric materials is rigorous only if the resonant frequencies of the cantilever in contact with the surface are identical.

Analysis of the relative electromechanical versus electrostatic contributions far from the resonances at high frequencies presents a more challenging problem. Intrinsic contributions (for example due to the electrocapillary effect at the tip–surface junction or capacitive cross-talk in cabling and the detector), can result in a shift of the nulling bias from the surface potential (for example the weak frequency dependence of the nulling bias), resulting in a non-zero effective piezoresponse even for non-piezoelectric materials. Figures 10(c) and (d) reveal the frequency dependence of the electromechanical contribution for PZT and SiO$_2$. While for most frequencies the effective
electromechanical response for PZT is ~2 orders of magnitude higher than that for SiO$_2$, the latter is non-zero, necessitating further studies.

7. Conclusions

The dynamic cantilever behaviour in vertical and lateral PFM is analysed using a combination of theoretical modelling and experimental 2D frequency–bias spectroscopy. The positions of the contact resonances are determined solely by the cantilever spring constant and the effective spring constant of the tip–surface junction. The relative contributions of the vertical and longitudinal electromechanical and local and non-local electrostatic contributions are analysed as a function of probe geometry and operation frequency. It is shown that quantitative electromechanical measurements require imaging at low tip biases or (for PFM spectroscopy) the use of high static loads and high spring constant cantilevers.

For longitudinal and lateral contributions, the onset of sliding friction at high frequencies or low indentation forces will minimize the transmission of surface vibrations to the tip. This suggests that lateral PFM imaging is optimal at relatively low frequencies, when the vibration transfer from the surface to the tip is effective. Conversely, vertical PFM is optimal at relatively high frequencies, when the longitudinal contributions are minimal.

Frequency–bias response diagrams are shown to be a convenient tool for analysis of image formation mechanisms in PFM. In particular, the frequency dependence of the nulling bias provides information on the electromechanical contribution to the signal and delineates frequency regimes for optimal quantitative electromechanical imaging. The electrostatic contribution to the PFM signal can be determined unambiguously from the slope of the data.

Acknowledgments

Support from ORNL SEED funding under Contract DE-AC05-00OR22725 is acknowledged (SVK). We also gratefully acknowledge the reviewer’s effort that has greatly contributed to the quality of this manuscript.

References

[1] Gruverman A, Auciello O and Tokumoto H 1998 Annu. Rev. Mater. Sci. 28 101
[2] Eng L M, Graffstrom S, Loppacher Ch, Schlaphof F, Trogisch S, Roolofs A and Waser R 2001 Adv. Solid State Phys. 41 287
[3] Alexe M and Gruverman A (ed) 2004 Nanoscale Characterization of Ferroelectric Materials (Berlin: Springer)
[4] Hong S (ed) 2004 Nanoscale Phenomena in Ferroelectric Thin Films (Dordrecht: Kluwer)
[5] Alexe M, Gruverman A, Harnagea C, Zakharov N D, Pignolet A, Hesse D and Scott J F 1999 Appl. Phys. Lett. 75 1158
[6] Roelofs A, Boettger U, Waser R, Schlaphof F, Trogisch S and Eng L M 2000 Appl. Phys. Lett. 77 3444
[7] Shin H, Hong S, Moon J and Jeon J U 2002 Ultramicroscopy 91 103
[8] Tybell T, Paruch P, Giamarchi T and Triscone J-M 2002 Phys. Rev. Lett. 89 097601
[9] Kalinin S V, Bonnell D A, Alvarez T, Lei X, Hu Z, Ferris J H, Zhang Q and Dunn S 2002 Nano Lett. 2 380
[10] Kalinin S V, Bonnell D A, Alvarez T, Lei X, Hu Z, Shao R and Ferris J H 2004 Adv. Mat. 16 795
[11] Terabe K, Nakamura S, Takekawa S, Kitamura K, Higuchi S, Gotot Y and Cho Y 2003 Appl. Phys. Lett. 82 433
[12] Kalinin S V, Rodriguez B J, Jesse S, Shin J, Baddour A P, Gupta P, Jain H, Williams D B and Gruverman A 2006 Microsc. Microanal. at press
[13] Ganpule C 2001 Nanoscale phenomena in ferroelectric thin films PhD Thesis University of Maryland, College Park
[14] Rodriguez B J, Gruverman A, Kingon A I, Nemanich R J and Ambacher O 2002 Appl. Phys. Lett. 80 4166
[15] Halperin C, Mutchnik S, Agronin A, Mulotski M, Urenski P, Salai M and Rosenman G 2004 Nano Lett. 4 1253
[16] Kalinin S V, Rodriguez B J, Jesse S, Thundat T and Gruverman A 2005 Appl. Phys. Lett. 87 053901
[17] Kalinin S V, Rodriguez B J, Jesse S, Thundat T, Grichko V, Baddour A P and Gruverman A 2006 Ultramicroscopy at press
[18] Hong S, Woo J, Shin H, Jeon J U, Park Y E, Colla E L, Setter N, Kim E and No K 2001 J. Appl. Phys. 89 1377
[19] Kalinin S V and Bonnell D A 2004 Nanoscale Characterization of Ferroelectric Materials ed M Alexe and A Gruverman (Berlin: Springer)
[20] Huey B D, Ramanujan C, Bobji M, Blendell J, White G, Szoszkiewicz R and Kulik A 2004 J. Electroceram. 13 287
[21] Likodimos V, Orikli X K, Pardi L, Labardi M and Allegrini M 2000 J. Appl. Phys. 87 443
[22] Harnagea C, Alexe M, Hesse D and Pignolet A 2003 Appl. Phys. Lett. 83 338
[23] Kalinin S V, Karapetian E and Kachanov A 2004 Phys. Rev. B 70 184101
[24] Karapetian E, Kachanov A and Kalinin S V 2005 Phil. Mag. 85 1017
[25] Rabe U, Kopycinska M, Hiserkorn S, Munoz-Saldana J, Schneider G A and Arnold W 2002 J. Phys. D: Appl. Phys. 35 2621
[26] Dinelli F, Castell M R, Ritchie D A, Mason N J, Briggs G A D and Kolossov O V 2000 Phil. Mag. A 80 2299
[27] Yamanaka K, Tsuji T, Noguchi A, Koike T and Mihara T 2000 Rev. Sci. Instrum. 71 2403
[28] This effect is partially considered in Abplanalp M 2001 Piezoresponse scanning force microscopy of ferroelectric domains PhD Thesis Swiss Federal Institute of Technology, Zurich
[29] Kalinin S V and Bonnell D A 2002 Phys. Rev. B 65 125408
[30] Eng L M, Güntherodt H-J, Schneider G A, Kopke U and Saldana J M 1999 Appl. Phys. Lett. 74 233
[31] Eng L M, Güntherodt H-J, Rosenman G, Skliar A, Oron M, Katz M and Eger D 1998 J. Appl. Phys. 83 5973–7
[32] Timoshenko S and Goodier J N 1951 Theory of Elasticity (New York: McGraw Hill)
[33] Rabe U, Janzer K and Arnold W 1996 Rev. Sci. Instrum. 67 3281
[34] Turner J A, Hiserkorn S, Rabe U and Arnold W 1997 J. Appl. Phys. 82 966
[35] Rabe U Atomic force acoustic microscopy Applied Scanning Probe Methods vol II, ed B Bhushan and H Fuchs (Berlin: Springer) at press
[36] Dienwiebel M et al 2005 Rev. Sci. Instrum. 76 043704
[37] Frederix P, Gullo M R, Akiyama T, Tonin A, de Rooij N F, Stauffer U and Engel A 2005 Nanotechnology 16 997
[38] Burnham N A, Chen X, Hodges C S, Matei G A, Thoresen E J, Roberts C J, Davies M C and Tendler S J B 2003 Nanotechnology 14 1
[39] Green C P, Lioe H, Cleveland J P, Proksch R, Mulvaney P and Sader J E 2004 Rev. Sci. Instrum. 75 1988
[40] Cleveland J P, Manns S, Bocek D and Hansma P K 1993 Rev. Sci. Instrum. 64 403
[41] Jesse S et al 2005 Preprint cond-mat/0509427
[42] Persson B N J 2004 Sliding Friction (Berlin: Springer)
[43] Mazeran P E and Loubet J L 1999 Tribol. Lett. 7 199
[44] Sarid D 1991 Scanning Force Microscopy (New York: Oxford University Press)
[45] Krotil H-U, Weilandt Th, Stifter Th, Marti O and Hild S 1999 Surf. Interface Anal. 27 341
[46] Kerssemakers J and De Hosson J Th M 1998 Surf. Sci. 417 281
[47] Rabe U, Turner J and Arnold W 1998 Appl. Phys. A 66 S277
[48] Turner J A and Wielen J S 2001 Nanotechnology 12 322
[49] Scherer V, Arnold W and Bhushan B 1999 Surf. Interface Anal. 27 578
[50] Reinstadtler M, Rabe U, Scherer V, Hartmann U, Goldade A, Bhushan B and Arnold W 2003 Appl. Phys. Lett. 82 2604
[51] This formula ignores fringe field and distributed electrostatic force will be actually larger. See Tiedke S and Schmitz T 2004 Ferroelectrics at Nanoscale: Scanning Probe Microscopy Approach ed M Alexe and A Gruverman (New York: Springer)
[52] Bdikin I K, Shvartsman V V, Kim S-H, Herrero J M and Kholkin A L 2004 Mater. Res. Soc. Symp. Proc. 784 C11.3