Effect of the intrinsic width on the piezoelectric force microscopy of a single ferroelectric domain wall

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Intrinsic domain wall width is a fundamental parameter that reflects bulk ferroelectric properties and governs the performance of ferroelectric memory devices. We present closed-form analytical expressions for vertical and lateral piezoelectric force microscopy (PFM) profiles of a single ferroelectric domain wall for the conical and disk models of the tip, beyond point charge and sphere approximations. The analysis takes into account the finite intrinsic width of the domain wall and dielectric anisotropy of the material. These analytical expressions provide insight into the mechanisms of PFM image formation and can be used for a quantitative analysis of the PFM domain wall profiles. The PFM profile of a realistic domain wall is shown to be the convolution of its intrinsic profile and the resolution function of PFM. © 2008 American Institute of Physics. [DOI: 10.1063/1.2939369]

I. INTRODUCTION

Piezoelectric force microscopy (PFM) has become the technique of choice for nanoscale imaging and characterization (see, e.g., Refs. 1–3), allowing for recent advances in PFM result interpretation.4 Now PFM methods are widely used for manipulation and tailoring of a ferroelectric domain structure (see, e.g., Refs. 5–7), study of ferroelectric domain growth dynamics,5 visualization and local characterization of capacitor structures,9 local polarization switching,10 polycrystalline and relaxor ferroelectrics,11 and size effects in ultrathin ferroelectric films.12 In order to interpret the experimental results of the PFM image of a ferroelectric domain wall with “sharp” tips (with high spatial resolution), one has to take into account the natural distribution of material properties such as the piezoelectric, dielectric, and elastic coefficients across the domain wall. For most of bulk and thin film ferroelectrics, the intrinsic wall width ranges from one to several lattice constants, as demonstrated by atomic force microscopy (AFM) and advanced transmission electron microscopy.13 A similar length scale has been found in polarization distribution over the ferroelectric film thickness determined from electron microscopy data.15 However, it was recently found that domain walls broaden on the surface compared with the bulk of a ferroelectric crystal.16,17 Furthermore, the presence of stresses, charge, or dipolar defects can result in elastic, dielectric, and electromechanical property gradients on length scales of 10–1000 nm. It is therefore essential to quantitatively understand the influence of the intrinsic property distribution across a wall on the observable PFM profile of the wall measured with sharp tips.

Recent results18,19 of domain wall PFM imaging as well as finite element modeling suggest that the intrinsic domain wall width has an effect on the measured PFM profile. Here we derive analytical expressions for vertical and lateral PFM profiles of finite-width 180° domain walls with predetermined polarization distribution, taking into account the contact and conical parts of the probe, as well as the dielectric anisotropy of the material. In realistic systems, both 180° ferroelectric and 90° ferroelastic domain walls appear. Ferroelastic domain wall intrinsic width is studied in Refs. 13 and 20. Using the relations between the values of wall width for 180° and 90° domains developed for the bulk systems,21 one could relate the wall widths in the bulk. However, the intrinsic width of a 180° domain wall near the surface may be strongly affected by mechanical and electrical boundary conditions,16 and hence the bulk relations21 should be modified.22 It should be noted that the difference in AFM and PFM profile widths, observed by Franck et al.20 for 90° domains in BaTiO3, may be related to different imaging mechanisms of AFM and PFM, determined by short-range contact forces and long-range electrostatic forces, respectively.

This paper is organized as follows. The basic principles of the PFM response calculation and electrostatic field structure of the probe are discussed in Sec. II. The relationship between the PFM profile of sharp domain walls and material
properties is analyzed in Sec. III for conic, contact, and effective point charge tip models. The influence of intrinsic width on domain wall PFM profile is considered in Sec. IV. Obtained results are discussed in Sec. V.

II. BASIC EQUATIONS

In the case of the strain piezoelectric coefficient $d_{klj}$ dependent only on lateral coordinates (a system may be considered uniform in the $z$-direction), the surface displacement vector $u_i(y)$ (measured PFM piezoresponce) is given by the convolution of an ideal image $d_{klj}(y-x)$ with the resolution function components $W_{ijkl}(x)$ (see Ref. 23). Since in many cases, the inhomogeneous distribution of piezoelectric coefficients are similar, e.g., for ferroelectrics they are determined by polarization distribution, hereinafter we introduce the inhomogeneous part of piezoelectric coefficients as $\beta(y-x)$, i.e., assume that $d_{klj}(y-x) = d_{klj} \beta(y-x)$. Here $d_{klj}$ are the piezoelectric tensor components of homogeneous media; the absolute value of function $\beta(y-x)$ is smaller than unity. Dielectric permittivity and elastic modules are regarded constant within the sample. In this approximation, components of the surface displacement below the tip can be written as follows:\(^\text{23}\)

$$u_i(y) = \int_{-\infty}^{\infty} dx_1 \int_{-\infty}^{\infty} dx_2 W_{ijkl}(-x_1,-x_2) d_{klj} \beta(y_1-y_1,y_2-y_2),$$

(1)

where the resolution function is introduced as

$$W_{ijkl}(x_1,x_2) = c_{kijn} \int_{0}^{\infty} dx_3 \frac{\partial G_{mn}(-x_1,-x_2,x_3)}{\partial x_i} E_i(x_1,x_2,x_3)$$

(2)

and $E_i$ is the component of the external electric field produced by the probe.

Here we calculate surface displacement below the tip located near the plain domain wall (see Fig. 1). For the infinitely thin isolated domain wall, Eq. (1) can be rewritten as

$$u_i^{\text{step}}(y_1) = \int_{-\infty}^{\infty} dx_1 \int_{-\infty}^{\infty} dx_2 W_{ijkl}(-x_1,-x_2) d_{klj} \delta(y_1-y_1) \delta(y_2-y_2).$$

(3)

Using the Delta function properties in Eq. (3), the displacement components for the arbitrary one-dimensional distribution $d_{klj} \beta(y_1)$, i.e., measured wall profile, is

$$u_i(y_1) = \frac{1}{2} \int_{-\infty}^{\infty} u_i^{\text{step}}(y_1-x) \frac{\partial \beta(x)}{\partial x} dx.$$  

(4)

Hereinafter $\beta(y_1)$ is the “intrinsic” domain wall profile (see Appendix A in Refs. 24 and 25) due to symmetry breaking, surface, or defect or strain effects on piezoelectric properties. Equation (4) is the main result that allows the calculation of a broadened wall profile if the profile $u_i^{\text{step}}$ of the infinitely thin domain wall is available.

Using decoupling approximation\(^\text{26,27}\) and resolution function theory,\(^\text{24}\) the piezoelectric response of the isolated 180° domain wall in the inhomogeneous electric field of the probe tip is

$$d_{33}^{\text{eff}} = \frac{u_1}{V} = g_{313} d_{31} + g_{333} d_{33} + g_{355} d_{15},$$

(5)

$$d_{33}^{\text{eff}} = u_2 = 0,$$

$$d_{33}^{\text{eff}} = \frac{u_3}{V} = g_{113} d_{31} + g_{133} d_{33} + g_{155} d_{15}.$$  

Here $V$ is the electric bias applied to the probe tip and Voigt matrix notations are used for piezoelectric tensor components. The explicit form of the tensorial functions $g_{ijkl}$ (and hence that of the PFM image) depends on the tip coordinates $\{y_1, y_2\}$, the domain wall intrinsic structure, and the electric field distribution of the probe.

Piezoelectric response in homogeneous electric field (flat capacitor geometry) is considered elsewhere.\(^\text{28}\) For the flat geometry the details of the tip structure are irrelevant since the bias is applied between the top and bottom electrodes and hence the electric field is uniform within the sample. In this case, the response is determined by three characteristic length scales: the thicknesses of the film, the thickness of the top electrode, and the radius of the tip-sample mechanical contact. For the case when the contact radius is much smaller than the first and second scales (approximation of point mechanical contact), one could use analytical expressions for the 180° domain wall piezoresponse derived in Ref. 28.

The electrostatic field produced by the tip includes the contributions from the conical part of the probe as well as the tip-surface contact area. The conical part can be approximated by a line charge (see, e.g., Refs. 29–31), and the contact area can be modeled by a disk touching the sample surface, as proposed in Ref. 18. Finally, the spherical part of the probe can be approximated by a point charge. Using the superposition principle, we represent the probe electrostatic potential, $\varphi$, as the sum of the effective line charge potential, $\varphi_L$, point charge potential, $\varphi_P$, and disk potential, $\varphi_D$:

$$\varphi(p,z) = \varphi_L(p,z) + \varphi_P(p,z) + \varphi_D(p,z),$$

(6)

where cylindrical coordinates $p = \sqrt{x_1^2 + x_2^2}$ and $z = x_3$ are introduced. Normalization in Eq. (6) requires $\varphi_L(0,0) + \varphi_P(0,0) + \varphi_D(0,0) = V$, corresponding to an ideal electrical contact between the tip and the surface. Under the condition $\varepsilon_e \ll \varepsilon_{11,33}$, typically valid for the majority of ferroelectrics with $\varepsilon_{11,33} \approx 100$ and $\varepsilon_e < 10$, we obtained expressions for a potential structure (see Appendixes B and C in Refs. 24 and 25):

FIG. 1. (Color online) Schematics of PFM measurement across 180° domain wall between domains with opposite polarizations $\pm P_3$. 

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Hereinafter $\kappa=\sqrt{\varepsilon_{33}/\varepsilon_{11}}$ is the effective dielectric constant, $\gamma=\sqrt{\varepsilon_{33}/\varepsilon_{11}}$ is the dielectric anisotropy factor, and $\varepsilon_0$ is the permittivity in free space. The conical part potential [Eq. (7a)] is modeled by the linear charge of length $L$ with a constant charge density $\lambda_L=4\pi\varepsilon_0\varepsilon_0 V/\ln(ctg^2\theta/2)$, where $\theta$ is the cone apex angle. An additional point charge potential [Eq. (7b)] is chosen to reproduce the conductive tip surface as closely as possible by the isopotential surface, $\varphi(\rho,z)=V$. Our numerical calculations show that the charge $q^*$ is located at the end of the line and that the distance $\Delta L$ from the surface is approximately equal to the disk radius $R_0$ and $q^*=4\pi\varepsilon_0\varepsilon_0 \varepsilon V\Delta L$ for a typical range of cone angles $\theta$. Contact area potential [Eq. (7c)] is modeled by the disk of radius $R_0$, with surface charge density $\sigma_d=V R_0/\sqrt{R_0^2-\rho^2}$, where $\rho$ is the radial coordinate [see Fig. 2(a)]. The vertical scale in Fig. 2(a) is compressed since $L<R_0$ for a real probe apex shape. It is clear from Fig. 2(b) that isopotential surface $\varphi(\rho,z)=V$ reproduces the conductive tip shape in the vicinity of the surface.

It is important for further consideration that under the conditions $\Delta L=R_0$, $\rho=R_0$, and $0<z$ the ratio of the conical terms (line + point charge) to the disk one can be estimated as $q^*/q_L=2\varepsilon_0 \varepsilon V/\ln(ctg^2\theta/2)$.

![Diagram](image)

**FIG. 2.** (Color online) (a) Tip-surface contact. (b) Corresponding isopotential lines near the sample surface at $\Delta L=R_0$, $\theta=\pi/9$, and $q^*=4\pi\varepsilon_0\varepsilon_0 \varepsilon V\Delta L$.

\[ \frac{\varphi_q+\varphi_L}{\varphi_D} \leq \frac{2\varepsilon_0 \varepsilon V}{\varepsilon_0 + \varepsilon \ln(ctg^2\theta/2)} \left( 1 - \frac{2\varepsilon_0 \varepsilon V}{\varepsilon_0 + \varepsilon \ln(ctg^2\theta/2)} \right)^{-1}. \] (7d)

So that it is small enough inside the sample under the typical conditions $\varepsilon_0 R_0 \leq 10^3$, $\theta=\pi/4$ because $\kappa\gg \varepsilon_0$, and $L/R_0 \ll \exp[(\varepsilon_0 + \varepsilon)/2\varepsilon_0]$. At the same time, the condition $\kappa\gg \varepsilon_0$ is necessary for the validity of Eqs. (7a)–(7c). Numerical simulations prove that the relative contribution of the conic potential ($\varphi_q+\varphi_L$) at $z=0$ is not more than 5% of the disk potential $\varphi_D$ (in agreement with the conclusion of Ref. 32). However we demonstrate below that the conic part may affect the piezoresponse saturation rate away from the domain wall.

It should be noted that the PFM profiles of domain walls for other tip models, such as sphere-plane, can be considered on the basis of the results above by summation or integration of the point charges, representing the tip. The PFM profiles of more sophisticated domain structures with ideal walls, such as periodic structures or cylindrical domains, were considered earlier for a spherical model of the tip. An effective point charge approach is developed in Ref. 33, neglecting the conic part contribution. The effective point charge approach uses only the point charge potential given by Eq. (7b), with $\Delta L \rightarrow h$, where the effective distance $h$ is proportional to the tip radius and depends on the tip model. In particular, the effective charge-surface separation is $h=2R_0/\pi$ for a disk model or $h=2\varepsilon_0 R_0 \ln[(\varepsilon_0 + \varepsilon)/2\varepsilon_0]/(\kappa - \varepsilon_0)$ for the sphere-plane one.

**III. THE INFLUENCE OF EXTRINSIC FACTORS ON THE WIDTH OF DOMAIN WALL PFM PROFILE**

The role of extrinsic factors on the broadening of a PFM image of a domain wall can be demonstrated by the PFM profile width of an infinitely sharp flat domain wall. In fact the width of the $d^W_{cr}(y)$ profile is an exactly extrinsic (i.e.,
measured) domain wall width as determined by the PFM object transfer function (OTF) finite half-width, which is defined by the tip size and geometry.

Using Eqs. (3), (5), and (7a)–(7d), displacement \( d_{32}^V \) at the distance \( y \) from an infinitely sharp domain wall was found to be as follows:

\[
d_{32}^V(y) = \frac{u^{\text{step}}(y)}{V} = \left( \frac{\varphi_0(0,0)}{V} g_{ijk}^S(y) + \frac{\varphi_0(0,0)}{V} g_{ijk}^G(y) \right) d_{kj}.
\]

Functions \( g_{ijk}^S(y, \Delta L) \), \( g_{ijk}^D(y, R_0) \), and \( g_{ijk}^L(y, \Delta L, L) \) could be presented via the universal function \( g_{ijk}(y, h_S) \) at \( s = q, D, L \) (see Appendixes B and C in Refs. 24 and 25). For a vertical piezoresponse, the nonzero displacement components are

\[
g_{313}^S(y, h_S) = (1 + \nu) f_{313}^p \rho_3^S \left( \frac{y}{C_{313}h_S} \right) - f_{333}^p \rho_3^S \left( \frac{y}{C_{333}h_S} \right),
\]

\[
g_{333}^S(y, h_S) = f_{333}^p \rho_3^S \left( \frac{y}{C_{333}h_S} \right) - f_{355}^p \rho_3^S \left( \frac{y}{C_{355}h_S} \right).
\]

For a lateral piezoresponse, the nonzero components are

\[
g_{113}^S(y, h_S) = (1 + \nu) f_{113}^p \rho_3^S \left( \frac{y}{C_{113}h_S} \right) - f_{133}^p \rho_3^S \left( \frac{y}{C_{133}h_S} \right),
\]

\[
g_{133}^S(y, h_S) = f_{133}^p \rho_3^S \left( \frac{y}{C_{133}h_S} \right) - f_{155}^p \rho_3^S \left( \frac{y}{C_{155}h_S} \right).
\]

The distances are \( h_L = h_L \approx \Delta L \) and \( d_D = R_0 \). Functions \( p_3^S(\xi) \) and \( p_3^G(\xi) \) represent the PFM image broadening of the ideal sharp domain wall. Their approximate expressions are

\[
\begin{align*}
p_3^S(\xi) &= \frac{\xi}{|\xi| + 1}, \\
p_3^G(\xi) &= \frac{1}{|\xi| + 1}.
\end{align*}
\]

\[
\begin{align*}
p_3^D(\xi) &= 2 \frac{\text{arctan}(\xi)}{\pi}, \\
p_3^G(\xi) &= \frac{2 \text{arctan}(1)}{\pi} \left( \frac{1}{|\xi| + 1} \right),
\end{align*}
\]

Here \( \nu \) is the Poisson ratio and \( s = L/D, L \). Constants \( C_{ijk} \) and \( f_{ijk} \) depend on the dielectric anisotropy factor \( \gamma \) only and are listed in Appendix D in Refs. 24 and 25. Constants \( f_{313} \) determine the value of the vertical response \( d_{32}^V(y) \) far from the wall at \( |y| \to \infty \). Constants \( f_{3ij} \) determine the value of the lateral response \( d_{ij}^S(y) \) at \( |y| \to 0 \). Constants \( C_{ijk} \) determine the effective width of the PFM image of the domain wall. It is obvious that when one of the terms \( f_{313} d_{kj} \) dominates the other, the half-width at half maximum (saturation level) for the lateral or vertical PFM profile will be \( C_{1jk}h_S \) or \( C_{3jk}h_S \), respectively. Material parameters and constants \( f_{3ij} \) and \( C_{ijk} \) for different ferroelectrics are listed in Tables I–III.

One can see from Eqs. (9a)–(9d) and (10a)–(10c) that the vertical response is zero at the center of the wall (at \( y = 0 \)) and saturates far from the wall (at \( |y| \to \infty \)), while the lateral PFM response is maximal in the center of the wall and tends to zero far from the wall. The vertical PFM response near an infinitely thin domain wall in PbTiO$_3$ as a function of the distance from the wall is shown in Fig. 3.

![FIG. 3.](image) (Color online) Vertical PFM response near the infinitely thin domain wall in PbTiO$_3$ as a function of the distance from the wall in (a) linear scale and (b) log-log scale. Solid, dashed, and dash-dotted curves are the full response, disk, and cone contributions, respectively. PbTiO$_3$ material parameters: \( \nu = 0.35, \kappa = 121, \gamma = 0.87, d_{333}^S = 117, d_{313}^L = 61, \) and \( d_{33}^D = -25 \) pm/V; ambient permittivity: \( e_0 = 1 \); tip characteristics: \( R_0 = L/20 \) nm, \( L = 2 \) \( \mu \)m, and \( \theta = \pi/9 \).
Since $R_0=\Delta L$, for the ratios $L/R_0\leq \exp[(\varepsilon_e+\kappa)/2\varepsilon_e]$, the relative contribution of the conic part to the vertical piezoresponse $d_{33}^C(y)$ is generally no more than 5%, while the disk contribution strongly dominates [compare solid, dashed, and dash-dotted curves in Fig. 3 and see Eq. (7d)]. The same is true for a lateral PFM response $d_{33}^D(y)$ (not shown). However, the conic part does affect the saturation rate of the piezoresponse from the domain wall due to the long-range character of the produced electric fields.

Since the contribution of the conic part is negligible in the vicinity of a domain wall $|y|<2R_0$ [see, e.g., Fig. 3(a)], the effective point charge approximation for a disk tip with $h=2R_0/\pi$ and $s=q$ is valid in the region $|y|<2R_0$ with satisfactory accuracy under the aforementioned condition $L/R_0\leq \exp[(\varepsilon_e+\kappa)/2\varepsilon_e]$ [see comments to Eq. (7d)]. Numerical simulations prove that the same inequality for the conic part length $L$ should be valid for the effective point charge approximation of the sphere-plane tip with curvature $R_0$.

The relative contribution of the conical part becomes comparable to the disk one under the condition $L/R_0\leq \exp[(\varepsilon_e+\kappa)/2\varepsilon_e]$. Figure 4 illustrates the region of tip geometric parameters where the disk part (i.e., contact) contribution into the piezoresponse dominates over the conic one. Obtained curves are well described by the dependence $\theta=\pi-2\arctg((1+L/R_0)e^{\Psi(\varepsilon_e+\kappa)})$ obtained directly from Eq. (7d), where the constant $\Psi=6-10$. Note that narrow “tapered” probes heavily favor local contribution to the signal, while probes with large opening angles favor nonlocal contribution.

IV. THE INFLUENCE OF INTRINSIC WIDTH ON DOMAIN WALL PFM PROFILE

The role of intrinsic factors on the broadening of the PFM image of a domain wall will now be considered for a domain wall with nonzero intrinsic (or natural) width. In order to consider the influence of intrinsic domain wall width analytically, we approximate the $d_{kl}$ distribution as the oblique step $\delta(x,b)=\{(x+b)-|x-b|\}/2b$, where $2b$ is the intrinsic width of the wall. Using Eqs. (4), (5), and (7a)–(7d), the piezoresponse $d_{33}^C(y,b)$ at the distance $y$ from the domain wall was found as follows:

$$d_{33}^{eff}(y,b) = \frac{u_y(y,b)}{V} + \frac{\varphi_1(0,0)}{V} g_{33}^L(y,b) + \frac{\varphi_1(0,0)}{V} g_{33}^D(y,b) + \frac{\varphi_1(0,0)}{V} g_{33}^R(y,b) \delta_{kj}.$$  \hspace{1cm} (11)

Functions $g_{33}^R(y,b,\Delta L)$, $g_{33}^D(y,b,R_0)$, and $g_{33}^L(y,b,\Delta L,L)$ could be presented via the universal function $g_{33}^m(y,b,h_m)$ at $m=q,D,L$. For the vertical piezoresponse, the nonzero components are

$$g_{313}^m(y,b,h_m) = (1 + \nu)f_{313}^{m} P_3^{m}\left(\frac{y}{C_{313}h_m}, \frac{b}{C_{313}h_m}\right)$$  \hspace{1cm} (12a)

$$g_{333}^m(y,b,h_m) = f_{333}^{m} P_3^{m}\left(\frac{y}{C_{333}h_m}, \frac{b}{C_{333}h_m}\right),$$  \hspace{1cm} (12b)

$$g_{355}^m(y,b,h_m) = f_{355}^{m} P_3^{m}\left(\frac{y}{C_{355}h_m}, \frac{b}{C_{355}h_m}\right).$$  \hspace{1cm} (12c)

For the lateral piezoresponse, the nonzero components are

$$g_{113}^m(y,b,h_m) = (1 + \nu)f_{113}^{m} P_1^{m}\left(\frac{y}{C_{113}h_m}, \frac{b}{C_{113}h_m}\right)$$  \hspace{1cm} (12d)

$$g_{133}^m(y,b,h_m) = f_{133}^{m} P_1^{m}\left(\frac{y}{C_{133}h_m}, \frac{b}{C_{133}h_m}\right),$$  \hspace{1cm} (12c)

$$g_{151}^m(y,b,h_m) = f_{151}^{m} P_1^{m}\left(\frac{y}{C_{151}h_m}, \frac{b}{C_{151}h_m}\right).$$  \hspace{1cm} (12d)

The distances are $h_q=h_L=\Delta L$ and $h_D=R_0$. Functions $p^q_1(\xi, \omega)$ and $p^D_1(\xi, \omega)$ represent the PFM image broadening of an oblique step domain wall,

$$p^q_1(\xi, \omega) = \frac{\ln(\xi+\omega)+1}{2\omega} + \frac{\ln(\xi-\omega)+1}{2\omega},$$  \hspace{1cm} (13a)

$$p^D_1(\xi, \omega) = \frac{\ln(\xi+\omega)+1}{2\omega} - \frac{\ln(\xi-\omega)+1}{2\omega},$$  \hspace{1cm} (13b)

$$p^L_1(\xi, \omega) = \frac{2}{\pi} \arctg(\xi+\omega) - \arctg(\xi-\omega) + \frac{1}{4\omega} \ln\left(\frac{\xi-\omega}{\xi+\omega+1}\right).$$  \hspace{1cm} (13c)
FIG. 5. (Color online) Vertical PFM response near the isolated domain wall in PbTiO3 as a function of the distance from the wall (a) with intrinsic width \( b = 10 \text{ nm} \) and different \( R_0 \) values (labels near the curves) and (b) with \( R_0 = 10 \text{ nm} \) and different intrinsic width \( b \) values (labels near the curves). A piecewise smooth distribution like the “oblique step” is shown schematically in the inset. Other parameters are the same as in Fig. 3.

Rather cumbersome expressions for conic part contributions \( p^D_{\text{conic}}(\xi, \omega) \) and \( p^D_{\text{contact}}(\xi, \omega) \) are listed in the end of Appendix D in Refs. 24 and 25.

Note that the first term in Eq. (13a) is independent of \( h_m \) and thus represents the “ideal image” of the domain wall, \( \beta(\xi, \omega) \), while the second term is related to the PFM object response function half-width. For small values, \( h_m \ll b \), and the second term is negligible near the wall center (i.e., at \( |y| \ll b \)). At the same time, the dependence of the PFM profile on the intrinsic width \( b \) is negligible far from the wall.

From Eqs. (12a)–(12d) and (13a)–(13d) the vertical response is zero at the center of the wall (at \( y = 0 \)) and saturates far from the wall (at \( |y| \to \infty \)), while the lateral PFM response is maximal at the center of the wall and tends to zero far from the wall. Note that at \( b = 0 \), Eqs. (12a)–(12d) and (13a)–(13d) derived for an oblique-like flat domain wall reduce to Eqs. (9a)–(9d) and (10a)–(10c) for an infinitely sharp domain wall, as anticipated. The vertical PFM response near the oblique domain wall in PbTiO3 as a function of the distance from the wall is shown in Fig. 5.

Numerical calculations prove that the relative contribution of the conic part to the effective piezoresponse \( e^{\text{eff}}_{33}(y) \) is negligible (about 1%–5%) in the case \( L/R_0 \ll \exp((\epsilon_r + \kappa)/2e_r) \), possible for typical ferroelectrics in air (since \( \kappa \gg \epsilon_r \)), while the disk contribution strongly dominates (similar to the case of infinitely sharp domain walls). However, the conic part affects the piezoresponse saturation rate. For a perfect tip-surface electric contact the contribution of the conical part is negligible in the vicinity of the domain wall \( |y| < 2R_0 \), providing validity for the effective point charge approximation with high accuracy under the aforementioned conditions.

It is clear from Fig. 4 that an intrinsic contribution to measured wall width becomes significant only for an intrinsic width \( b > R_0/2 \). Moreover, both analytical results [Eqs. (12a)–(12d) and (13a)–(13d)] and numerical simulations for different intrinsic domain wall profiles \( \beta(x) \) lead to the same conclusion: the intrinsic domain width effect on the PFM response becomes negligible at \( 2b < h_m \) over all ranges of available material parameters. This rather general result, along with the estimation of \( h_m \sim 5–50 \text{ nm} \) (valid for standard conductive tips depending on their curvature and experiment geometry) and the inequality \( b < (2–5) \text{ nm} \) (typical for bulk perovskites such as PbTiO3 or BaTiO3,34 LiTaO3,16 and Rochelle salt18), corroborates the proposed tip calibration procedure elaborated in Ref. 35 for the effective point charge approach and an infinitely sharp domain wall approximation.

The opposite situation \( b \gg 10 \text{ nm} \) may be realized in LiNbO3 (Ref. 18) and organic polar materials. Also one may suspect \( h_m < 5 \text{ nm} \) for atomic or ultrasharp tips with small curvature \( R_0 \). Under the condition \( b > h \) both effective point charge-surface separation \( h \) and intrinsic width \( b \) should be taken into account for an accurate fitting of the domain wall PFM profiles. The aforementioned tip calibration procedure should also be modified, as discussed below.

V. DISCUSSION

Closed-form analytical expressions (11), (12a)–(12d), and (13a)–(13d) for vertical and lateral PFM profiles of the finite-width domain wall are derived. They take into account the conical and contact parts of the probe and the material dielectric anisotropy \( \gamma \). Similar to the case of an infinitely sharp domain wall, under the condition \( L/R_0 \ll \exp((\epsilon_r + \kappa)/2e_r) \) the contribution of the conical part is negligible in the vicinity of the domain wall, providing validity for the effective point charge approximation with high accuracy.

In the effective point charge approximation of the tip,36 electric field, and dielectric anisotropy \( \gamma = 1 \), the vertical piezoresponse at a distance \( y \) from the oblique domain wall \( \beta(y, b) = (|y + b| - |y - b|)/2b \) located at \( y = 0 \) acquires the following simplest form:

\[
d^{\text{eff}}_{33}(y, b, h) = \left[ -\left( \frac{1}{4} + \nu \right) d_{31} + \frac{3}{4} d_{33} + \frac{d_{15}}{4} \right] \frac{|y + b| - |y - b|}{2b} - \frac{d_{15} 3h}{4 8b} \ln \left( \frac{4|y + b| + 3h}{4|y + b| + 3h} \right) - \left( \frac{1}{4} + \nu \right) d_{31} \frac{3}{4} d_{33} \times \frac{h}{8b} \ln \left( \frac{4|y - b| + h}{4|y + b| + h} \right) \right].
\]

(14)

Here the first term is the ideal image \( \beta(y, b) \) of the domain wall intrinsic profile. Equation (14) can be used for two-parametric domain wall profile fitting and tip calibration, where the effective point charge-surface separation \( h \) and intrinsic width \( b \) are fitting parameters.

When the intrinsic width \( b \) is several times smaller or greater than the effective point charge-surface separation \( h \), Eq. (14) can be further simplified to...
\[ d_{33}^{\text{eff}}(y, b, h) = - \beta(y, b) \left[ \frac{|y| + b}{|y| + b + h/4} \left( \frac{1}{4} + \nu \right) d_{31} + \frac{3}{4} d_{33} \right] + \frac{d_{15}}{4} \frac{|y| + b}{|y| + b + h/4} \].

(15)

Numerical calculations prove that Eq. (15) is approximately valid for an arbitrarily smooth domain wall profile \( \beta(y, b) \), with characteristic intrinsic width \( b \) [e.g., \( \text{for tanh}(y/b) \) or \( 1 - \exp(-|y|/b) \text{sgn}(y) \)]. Thus, for a given \( \beta(y, b) \) we could fit the measured domain wall profile \( d_{33}^{\text{eff}}(y) \) and extract \( h \) and \( b \) values. The essential progress of closed-form expressions (11), (12a)--(12d), and (13a)--(13d) and especially approximation (15) in comparison to a more rigorous numerical fitting of experimental data demonstrated earlier\(^{18} \) consists in the possibility of a simple and unique analytical interpretation of available experimental data. The disadvantage of Eq. (15) is its insufficiency for an accurate quantitative domain wall profile reconstruction, while qualitative analyses can be easily performed.

On the other hand, the first term in Eq. (15) is the ideal image \( \beta(y, b) \) of the domain wall intrinsic profile. For \( b \to 0 \) the second bracket is exactly the absolute value of the PFM response of an infinitely sharp domain wall\(^{37} \) and the function \( \beta(y, b) \to \text{sign}(y) \), as anticipated. Actually, Eq. (15) is the product of an intrinsic (structural) factor and a pseudoelectric factor related to OTF features and intrinsic width \( b \) superposition. Note that we could not neglect \( b \) in the immediate vicinity of the domain wall (\( |y| \ll b \)) even in the case \( b \ll h \). Thus, the extrinsic factor appears naturally broadened with intrinsic width \( b \), proving that intrinsic and extrinsic factors cannot be easily separated.

Within the effective point charge approximation, both intrinsic width \( b \) and effective tip parameter \( h \) can be extracted from the measured piezoelectric profile slope in the immediate vicinity of the domain wall (i.e., at \( |y| \ll b \)), and the piezoelectric saturation rate far from the wall (i.e., at \( |y| \gg b + h \)) can be determined since expansions exist,

\[ d_{33}^{\text{eff}}(y, b, h) \approx - \beta(y, b) \left[ \frac{d_{15}}{4} \frac{1}{b + h/4} + \left( \frac{1}{4} + \nu \right) d_{31} + \frac{3}{4} d_{33} \right], \quad |y| \ll b, \] (16a)

\[ d_{33}^{\text{eff}}(y, b, h) \approx \left[ \frac{1}{4} + \nu \right] d_{31} + \frac{3}{4} d_{33} \left( 1 - \frac{h}{4y} \right), \quad |y| \gg b + h. \] (16b)

Equations (16a) and (16b) are valid quite satisfactorily for an arbitrarily smooth domain wall profile \( \beta(y, b) \), with characteristic intrinsic width \( b \). So, for materials with known piezoelectric coefficients \( d_{ij} \) the procedure for \( h \) and \( b \) determination is as follows:

1. At the first step the effective tip parameter \( h \) is determined from the piezoelectric profile fitting far from the wall by Eq. (16b) since \( d_{33}^{\text{eff}}(y) = d_{33} - d_{15} h/4 \).

2. At the second step the tangential slope \( \alpha \) near the wall can be determined using least squares method and Eq. (16a) since here \( d_{33}^{\text{eff}}(y) = \alpha y \). Then the intrinsic width \( b \) can be obtained from the slope \( \alpha \) as a solution of quadratic equation.

To summarize, obtained analytical expressions provide insight into the mechanisms of domain structure PFM image formation. Namely, the PFM profile of a realistic domain wall is the complex convolution of its intrinsic profile and extrinsic factors related to the nonlocality of the PFM resolution function; it could not be reduced to their product even in the simplest cases. This result distinguishes PFM imaging from the “far-field” methods, where a typical diffraction pattern represents the product of structural and scattering factors.

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25See EPAPS Document No. E-JAPIAU-103-074812 for details of electric and elastic fields calculations. For more information on EPAPS, see http://www.aip.org/epaps/numbering.html
36Starting from the tip model of point charge series, numerical fitting of the domain wall profiles in LiNbO3 and Pb(Zr,Ti)O3 converged to the single charge both in air and liquid ambient.