Theoretical Model of SPM-tip Electrostatic Field Accounting for Dead Layer and Domain Wall

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***Abstract*—A new theoretical approach for the determination of the electric field distribution in the ferroelectric/dielectric system with the presence of the SPM tip is presented. For the analytical solution of the problem some simplifications are introduced (initial model statement has only a numerical solution). Namely, the smallness of the domain wall thickness in the comparison with a domain size and high value of the ferroelectric dielectric permittivity are used. The developed approach allows us to obtain explicit formulas for the polarization and electric field intensity. The calculation and analysis of the tip capacitance as a function of the distance from the ferroelectric interface demonstrate that the presence of charges at the domain wall results in the difference in obtained values of 30% in comparison with the widely used dielectric model [1].**

***Index Terms*—ferroelectric domain, domain wall, dead layer, electrostatic field, SPM tip.**

I. Introduction

The employment of the ferroelectric domain structures in memory and optical devices requires to calculate accurate electric field distribution. In literature it is possible to find a number of papers focused on the solution of this problem [2]–[4]. However, there is no explicit formulas describing a full physical picture. At the present moment, the most widely used approach for the Scanning Probe Microscope (SPM) tip-induced field is based on the consideration of the point source at the interface of two dielectrics [5], [6]. Such a model obviously does not account for physical features of the ferroelectric materials. For a proper description of the electric field distribution it is necessary to take into consideration the following factors: (i) non-spherical form of the tip, (ii) the presence of a "dead" or passive layer at the ferroelectric/dielectric interface, (iii) the presence of the domain wall, and finally (iv) finite thickness of the ferroelectric layer and influence of the substrate. The form of the domain is considered as semi-ellipsoid [7]. In this work, we have investigated the impact of the aforementioned factors on the modeling of the electric field distribution. The pure electrostatic model is presented, i.e. the influence of elastic stresses is not considered (in spite of the necessity to examine this effect as was reported in [2]–[5]) The model generalization accounting for the piezoelectric effect will be published soon.

II. The approach

The theoretical description of the electric field distribution in the transition layer (to which domain wall and "dead" layers can be related) is quite complicated and no known complete solution exists. It is worth mentioned that the additional complexity arises due to the absence of any established theory of charge distribution in this kind of layers. For a solution of this problem we introduce approximate boundary conditions. The main quantities describing the electric field are electric potential U, electric field intensity $\vec{E} = -\nabla U$, polarization $\vec{P}$ and electric displacement $\vec{D} = \varepsilon_0\vec{E} + \vec{P}$, where ε_0 is the vacuum permittivity.

A. One-dimensional case

As a starting point, let us consider the simplest one-dimensional problem (i.e. we deal with scalar quantities). Assume that the transition layer lies in the interval $[-l/2, l/2]$. For any dependence between the polarization and the electric field intensity the last one should follow Poisson's equation

$$\varepsilon_0 \frac{\mathrm{d}E(z)}{\mathrm{d}z} = -\frac{\mathrm{d}P(z)}{\mathrm{d}z} + \rho(z), \qquad (1)$$

where $\rho(z)$ is the unknown volume charge charge density ($\rho(z) = 0$ outside the layer). After integration of both sides of (1) over z in the range from $-l/2$ to $l/2$ we can write

$$\varepsilon_0(E_+ - E_-) = \sigma. \qquad (2)$$

Here and below indexes + and - relate to corresponding quantities specified at $z = \pm l/2$. It is worth mentioned that in (2) a transition from the volume charge density to an effective surface charge density σ is performed

$$\sigma = \int_{-l/2}^{l/2} \rho(z) z \mathrm{d}z + P_- - P_+. \qquad (3)$$

At the same time, if we preliminary multiply (1) by z and then integrate over the transition layer, it is possible to derive following expression for the potential

$$\varepsilon_0(U(+0) - U(-0)) = \tau, \qquad (4)$$

where

$$\tau = \int_{-l/2}^{l/2} z \left(\rho(z) - \frac{\mathrm{d}P(z)}{\mathrm{d}z}\right) \mathrm{d}z. \qquad (5)$$

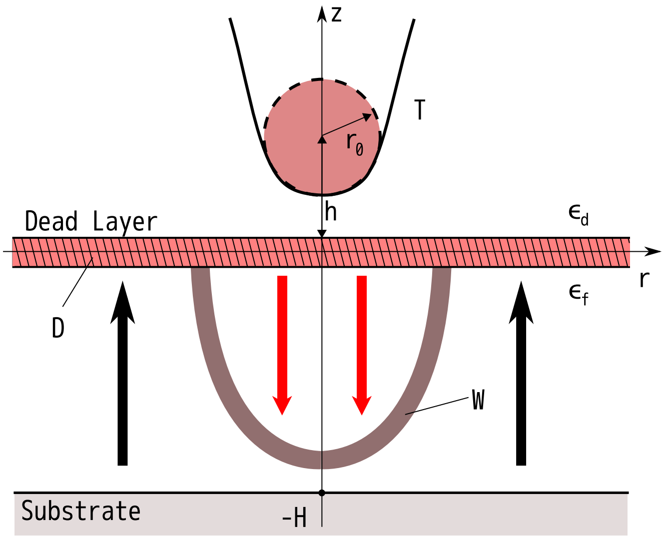

Fig. 1. Schematic representation of the SPM tip/dead layer/ferroelectric film/substrate or/ bottom electrode configuration. The arrows inside the structure indicate the spontaneous polarization directions.

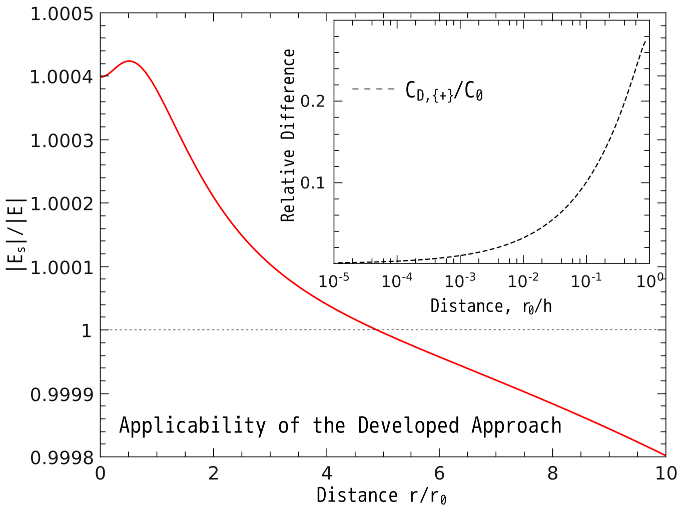

Fig. 2. The dependence of the dimensionless coordinate on the ratio between the electric field absolute value obtained with the precise boundary conditions (6) $|E|$ and that defined by the developed approach $|E_\mathrm{s}|$. The calculation is performed for $z = r_0/2$. Inset: the relative difference between obtained results using a new approach and dielectric model [1] in the case of the ferroelectric spontaneous polarization vector pointing up.

The boundary conditions (2-5) are precisely defined and allow significantly simplify the mathematical problem statement. That is, instead of finding the unknown functions $P(z)$ and $\rho(z)$ we suggest to define two phenomenological constants – the effective surface charge density σ and the potential shift τ. The potentials obtained by means of $P(z)$,$\rho(z)$ and σ,τ are identical with a proper choice of these constants. In other words, application of the boundary conditions (2-5) do not result in the sacrifice of accuracy.

B. Account of the high dielectric constant of ferroelectrics

Having obtained information about the general structure of the considered layer boundary conditions, we pass to the description of the system dielectric/ferroelectric with the interface at $z = 0$ (see Fig. 1). As an example of the model applicability, the tip end is represented by a sphere with radius r_0. The dielectric film of thickness H having an underlying conducting substrate. The tip end is away from the ferroelectric interface by the distance h. Quantities related to dielectric and ferroelectric have index d and f, respectively. In this notation the boundary conditions at $z = 0$ can be written as

$$U_\mathrm{f}(r,z)|_{z=0} = U_\mathrm{d}(r,z)|_{z=0}, \quad \varepsilon_\mathrm{f} \left.\frac{\partial U_\mathrm{f}(r,z)}{\partial z}\right|_{z=0} = \varepsilon_\mathrm{d} \left.\frac{\partial U_\mathrm{d}(r,z)}{\partial z}\right|_{z=0}. \tag{6}$$

We introduce a small parameter $s = \varepsilon_d/\varepsilon_f$. For conventional ferroelectric materials this parameter varies in the range of $10^{-4}\div10^{-2}$ [8]. According to [9] the electric field potential can be expressed by the following power series over s

$$U_{\mathrm{f}\{\mathrm{d}\}}(r,z) = \sum_{i=0}^{\infty} U_{\mathrm{f}\{\mathrm{d}\},i}(r,z) s^i. \tag{7}$$

The substitution of (7) in (6) and equalization of the coefficients with the same power of s leads to the system of equation. From this system, we can define $U_{\mathrm{f}0} = 0$ and boundary conditions for the first nonzero terms of the series

$$U_{\mathrm{d},0}(r,z)|_{z=0}, \quad \left.\frac{\partial U_{\mathrm{f},1}(r,z)}{\partial z}\right|_{z=0} = \left.\frac{\partial U_{\mathrm{d}0}}{\partial z}\right|_{z=0}. \tag{8}$$

From (8) follows that for small s the electric field distributions in the dielectric and ferroelectric can be considered independently from each other. This fact significantly simplifies the calculations. To be more precisely, from the first of conditions (8) the electric field in the dielectric is found. Further, the electric field in the ferroelectric film is defined by using the known value of the electric field intensity at the interface. The comparison of the electric field absolute value for precise model $|E|$ (i.e. based on (6)) and that obtained by the suggested scheme $|E_\mathrm{s}|$ is demonstrated in Fig. 2. One can see a perfect agreement between the aforementioned approaches, i.e. precise model can be reproduced by the simplified theory with enough accuracy.

C. Mathematical statement of the problem

The described simplifications allow us to formulate the statement of the problem for electric field distribution determination in the system depicted in Fig. 1. The dielectric and ferroelctric potentials should obey Laplace equation

$$\Delta U_\mathrm{f}(r,z) = 0, \quad \Delta U_\mathrm{d}(r,z) = 0, \tag{9}$$

and the boundary conditions

$$
\begin{aligned}
&(U_\mathrm{d}(r,z) - U_\mathrm{f}(r,z))|_{z=0} = \tau_\mathrm{int}(r), \quad U_\mathrm{d}|_T(r,z) = V,\\
&\left.\left(\varepsilon_\mathrm{d}\frac{\partial U_\mathrm{d}(r,z)}{\partial z} - \varepsilon_\mathrm{f}\frac{\partial U_\mathrm{f}(r,z)}{\partial z}\right)\right|_{z=0} = \sigma_\mathrm{int}(r),\ U_\mathrm{f}|_{z=-H} = 0,\\
&\left[\frac{\partial U_\mathrm{f}(r,z)}{\partial \mathrm{n}}\right]_\mathrm{W} = \sigma_\mathrm{D}(r,z), \quad [U_\mathrm{f}(r,z)]_\mathrm{W} = \tau_\mathrm{D}(r,z). \quad (10)
\end{aligned}
$$

Here T is the tip surface area, V the tip-surface potential, W the domain wall, and n normal to W. Symbols $[A]_\mathrm{W}$ mean discontinuity of a quantity A while approaching W by normal from different sides [10]. Effective surface charge density at the dielectric/ferroelctric interface $\sigma_\mathrm{int}(r)$ as well as for the "dead" layer $\sigma_\mathrm{D}(r)$ can be defined by (3).

D. Solution of the problem

At first we consider the case when substrate is excluded from a consideration, i.e. the ferroelectric layer is assumed infinite. In this case the potential of the system can be written as

$$
U_\mathrm{BULK}(r,z) = U_\mathrm{TIP}(r,z) + U_\mathrm{W}(r,z) + U_\mathrm{D}(r,z), \quad (11)
$$

where U_TIP is the potential contribution of the SPM tip, U_W potential of domain wall, and U_D dead layer contribution.

For a finding of the explicit expressions for potentials in the case of the sphere-shape tip, let us introduce bispherical coordinates (α,β) by the use of the chain of relations

$$
r = \frac{c\sin\alpha}{\cosh\beta - \cos\alpha}, \quad z = \frac{c\sinh\beta}{\cosh\beta - \cos\alpha}. \quad (12)
$$

where c is the scaling multiplier. In this notations the influence of the SPM tip on dielectric and ferroelectric layers is

$$
\begin{aligned}
U_\mathrm{TIPd} &= V_\mathrm{sph}\sum_{i=0}^{\infty}\frac{s\sinh\left(i+\frac{1}{2}\right)\beta + \cosh\left(i+\frac{1}{2}\right)\beta}{s\sinh\left(i+\frac{1}{2}\right)\beta_0 + \cosh\left(i+\frac{1}{2}\right)\beta_0}\times\\
&\times\exp\left[-\left(i+\frac{1}{2}\right)\beta_0\right]P_\mathrm{n}(\cos\alpha),\\
U_\mathrm{TIPf} &= V_\mathrm{sph}\sum_{i=0}^{\infty}\frac{\exp\left[\left(i+\frac{1}{2}\right)(\beta-\beta_0)\right]P_\mathrm{n}(\cos\alpha)}{s\sinh\left(i+\frac{1}{2}\right)\beta_0 + \cosh\left(i+\frac{1}{2}\right)\beta_0}
\end{aligned}
\quad (13)
$$

where $V_\mathrm{sph} = V\sqrt{2(\cosh\beta - \cos\alpha)}$ and P_n are Legendre's polynomials of n-th order [6], [9]. Ferroelectric/dielectric interface $z = 0$ in bispherical-coordinate system is defined as $\beta = 0$. The tip surface is determine by $\beta = \beta_0$ with

$$
\beta_0 = \mathrm{arcosh}\left(\frac{h}{r_0}\right), \quad h = c\coth\beta_0, \quad r_0 = \frac{c}{\sinh\beta_0}. \quad (14)
$$

It should be noted that the developed approach is applicable for a hyperbolic-shape tip [10], but this is out of the scope of this work. Corresponding contribution of the "dead" layer potential in dielectric and ferroelectric is given by

$$
\begin{aligned}
U_\mathrm{Dd} &= \sum_{i=0}^{\infty}\frac{p_\mathrm{i}\left[\sinh\left(i+\frac{1}{2}\right)\beta - \tanh\beta_0\cosh\left(i+\frac{1}{2}\right)\beta\right]}{\left(i+\frac{1}{2}\right)(\varepsilon_\mathrm{d}+\varepsilon_\mathrm{f}\tanh\beta_0)/P_\mathrm{n}(\cos\alpha)},\\
U_\mathrm{Df} &= \sum_{i=0}^{\infty}\frac{p_\mathrm{i}\exp\left[\left(i+\frac{1}{2}\right)\beta\right]\tanh\beta_0 P_\mathrm{n}(\cos\alpha)}{\left(i+\frac{1}{2}\right)(\varepsilon_\mathrm{d}+\varepsilon_\mathrm{f}\tanh\beta_0)}
\end{aligned}
\quad (15)
$$

where the coefficient p_i depends on the direction of spontaneous polarization in ferroelectric layer and domain size.

Based on the derived expressions (13),(15) we undertook a comparison of the SPM tip capacitance with and without account of "dead" layer contribution (U_Dd) to U_BULK. The calculation results are demonstrated in Fig. 3. Due to the difference in obtained results of $\sim 30\%$ (see Fig. 2, inset) it is become obvious that the account of the "dead" layer influence on the total electric field of the system is extremely significant.

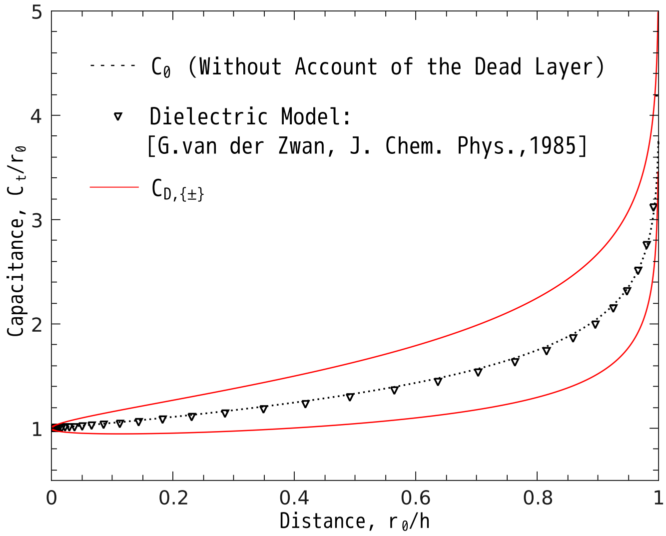

Fig. 3. The dimensionless capacitance (r_0 is the SPM-tip radius) as a function of the distance from the ferroelectric interface h. The signs of $C_{\mathrm{D},\{\pm\}}$ correspond to the ferroelectric spontaneous polarization direction.

For the ferroelectric layer with a finite thickness using the mirror-image method [6] we obtain

$$
\begin{aligned}
U_\mathrm{TOTAL}(r,z) &= U_\mathrm{BULK}(r,z) + \sum_{i=1}^{\infty}U_\mathrm{BULK}(r,z-4Hi)+\\
&+U_\mathrm{BULK}(r,-z-4Hi) - U_\mathrm{BULK}(r,z+2H-4Hi)-\\
&-U_\mathrm{BULK}(r,-z+2H+4Hi).
\end{aligned}
\quad (16)
$$

Equation (16) is much more simple than the one obtained in [3]–[5] by employing the Fourier transformation.

In order to describe the domain wall induced field and the electric field of hyperbolic-shape tip, the degenerate ellipsoidal coordinate system is employed [6]. As an example of this coordinate system application we consider only the determination of electric field distribution induced by the hyperbolic-shape SPM tip. The degenerate ellipsoidal coordinate system (ξ, η) for a prolate ellipsoid of rotation is given by

$$r = a \sinh \xi \sin \eta, \quad z = a \cosh \xi \cos \eta, \tag{17}$$

where a is the scaling multiplier. The surface of the tip is defined by hyperboloid $\eta = \eta_0$. Parameter a has a sense of focal distance of considered hyperboloid. After simple calculations we obtain expression for the electric field distribution in the dielectric medium

$$U_{\mathrm{d}} = V \frac{Q_0(\eta)}{Q_0(\eta_0)}. \tag{18}$$

Here $Q_0(\eta)$ is the zeroth-order Legendre function of the second-kind. A thorough analysis of (13) and (18) allows us to conclude that the potential of the hyperbolic-shape tip exceed the sphere-shape tip potential for any point for any point of the system being investigated (Fig. 1).

III. Conclusion

We have suggested a new approach for the determination of the electric field distribution in the ferroelectric/dielectric system with the presence of the SPM tip. The electric field in the dielectric can be obtained assuming the absence of the interface potential. Meanwhile, the distribution of the electric field in the ferroelectric layer is possible to derive using the normal component of the field intensity at the interface. The calculation error of the described approach is $O(\varepsilon_{\mathrm{d}}/\varepsilon_{\mathrm{f}})$. On the base of the proposed model we have made several important conclusions. Namely, the contributions of the domain wall and "dead" layer the total electric field of the system are significant ($\sim 30\%$) and should be taken into consideration. The influence of the substrate on the potential distribution is decreased as $\sim 1/(2H)^2$ with growing ferroelectric layer thickness. In other words, by means of our model the position where the substrate contribution is negligible can be defined.

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