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Scanning probe microscopy of well-defined periodically poled ferroelectric domain structure

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We analyze and determine the factors governing the contrast in contact mode atomic force microscopy of domain-structured ferroelectric crystals. The analysis is applied to measurements conducted on $KTiOPO_4$ crystals with artificially created well-defined domain structure. It is found that the amplitude contrast is due to difference in the work functions of the antiparallel domains. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1456967]

Scanning probe microscopy of ferroelectric domains uses the basic properties of ferroelectrics, such as their piezoelectric behavior and the presence of surface electrostatic field associated with the permanent built-in electric polarization. The two general atomic force microscopy (AFM) modes for ferroelectric domain imaging—contact and noncontact—were introduced by Franke[1] and Saurenbach.[2] In most papers to date, the electromechanical behavior (piezoresponse) of differently oriented domains is regarded as the only cause for the domain contrast in the contact mode of operation.[3–5] In the noncontact mode the domain image is asserted to be dominated by the effect of different electric fields emerging from the surface of the ferroelectric domains.[2,3] In one case, Hong *et al.*[6] have claimed that different electrical properties of the domains and not the piezoresponse, is what governs the domain contrast in the contact mode.

The piezoresponse mode of the AFM is based on the detection of the local vibration of the ferroelectric sample caused by the converse piezoelectric effect when an external ac voltage is applied between the sample and the conducting AFM tip that are in contact. The cantilever oscillates together with the sample, and the amplitude or the phase of its motion are measured using lock-in techniques. Usually the product of the amplitude and the phase is called piezoresponse signal. It is generally assumed[4,5] that the phase of the piezoresponse signal depends on the sign of the piezoelectric coefficient (and on the polarization direction) and changes by 180° in antiparallel domains. This means that opposite orientation of polarization leads to contrast in the piezoresponse image. In this approach the vibration amplitude provides information about the absolute value of the piezoelectric coefficient and consequently *should not depend on the polarization direction*. This is true in the case when the ferroelectric domains extend throughout the sample thickness.

In ferroelectric thin films, the piezoresponse signal provides integral information about all the randomly polarized grains distributed throughout the film thickness.[4] Therefore, the presence of both opposite polarized grains, and grains with polarization vector not perpendicular to the surface plane will change the contrast relative to that obtained when all the grains are polarized in the same direction, perpendicular to the sample surface. The role of the piezoresponse in the domain imaging was emphasized[7,8] for ferroelectric thin films. However, Likodimos[9] reported on a well-defined contrast between antiparallel domains in TGS single crystal observed in the amplitude signal.

The objective of the present work is to determine the factors governing the domain contrast in the contact AFM mode and to clarify the above-mentioned controversies. For this purpose we have measured a ferroelectric crystal with known properties and *artificially created well-defined domain structure*. The measurements were performed both on the Z-polar and Y-nonpolar faces of the ferroelectric. This allowed us to separate the influence of the electrostatic forces, which may occur only on the polar face, from the piezoresponse effect possible for both Z and Y faces.

We have used a 0.46-mm-thick $KTiOPO_4$ (KTP) ferroelectric crystal related to $mm2$ group. It is well known[10] that structural channels propagating through the KTP crystal framework parallel to the polar z-crystallographic axis provide fast potassium ionic transport resulting in very high ionic conductivity ($\sigma \sim 10^{-6}$ S/cm[10]). Ferroelectric periodic domain structure (period of 24.7 μm, the domains extend throughout the sample thickness) was produced by electrical poling as described in detail in the past.[11] The measurements were conducted using a commercial AFM (Autoprobe CP, Thermomicroscopes, Inc.). A heavily doped silicon cantilever with a spring constant of ~0.5 N/m was used for the contact measurements; the nominal radius of the tip was ~10 nm.

Optical microscopy image of the tailored strip-like domain configuration is shown in Fig. 1 (top view). Two faces of the sample were studied. Figures 2(a) and 2(b) show EFM amplitude contact mode ferroelectric domain images of thoroughly polished polar Z and nonpolar Y faces of the KTP sample, respectively. In the case of Z-face ac voltage $V = V_{ac} \sin \omega t$ with $V_{ac} = 15$ V, $\omega = 1.5$ kHz was applied in the polar Z direction of the crystal between the AFM tip and the bottom electrode. For the nonpolar face the voltage was applied along the Z direction, while the tip was scanning on the

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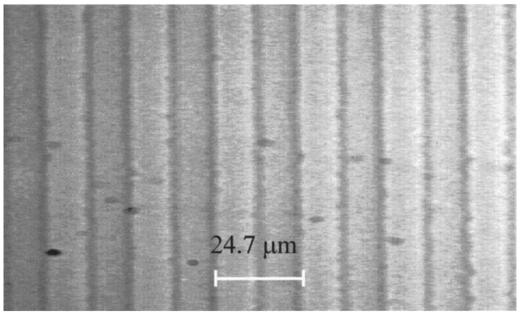

FIG. 1. Optical image (top view) of the periodic domain structure of the KTP crystal after etching.

Y surface plane as shown in Fig. 3. It is clearly observed that the periodic contrast in Fig. 2(a) has a period coinciding exactly with that of the fabricated domain structure (Fig. 1), while in Fig. 2(b) there is no domain contrast at all.

The sample topography imaged simultaneously using contact AFM was found to be featureless for both cases (not shown). Hence, the domain structure does not affect the sample topography image. Since the domains in the measured crystal extend throughout the sample, in the common (piezoresponse) interpretation of images like the one in Fig. 2(a) no domain contrast should be observed. This is because the absolute value of the piezoelectric coefficient is the same for the adjacent antiparallel domains. Moreover, in both the polar and the nonpolar cases the corresponding piezoelectric coefficients in KTP crystals (d_{32}) and (d_{33}) are of the same order of magnitude.[12] Therefore, the contrast difference in the EFM signal for these measurements [Figs. 2(a) and 2(b)] supports our hypothesis that the contrast in the contact mode images is not due to different piezoresponse of antiparallel domains.

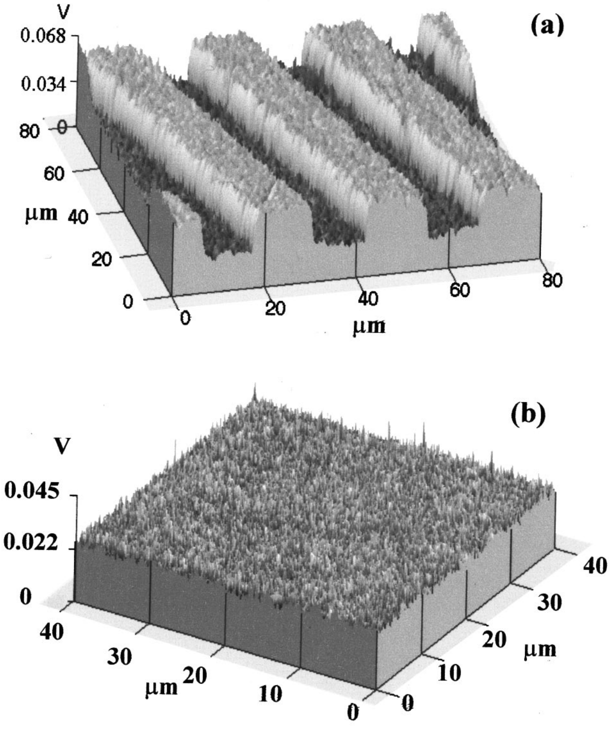

FIG. 2. Contact EFM image of the domain grated structure of $KTiOPO_4$ crystal: (a) polar face, (b) nonpolar face.

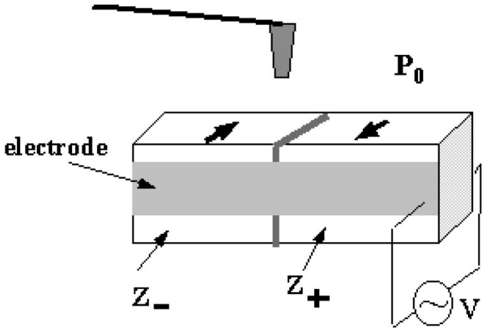

FIG. 3. Schematic of the nonpolar face EFM measurement setup.

Another possible cause for the domain contrast is a difference in the electrical properties of antiparallel domains. The screening of the depolarization field in ferroelectrics occurs by surface band bending.[13] In comparison with the known phenomenon of band bending in conventional semiconductors the band bending in ferroelectrics is asymmetric. Near the positive polar Z^+ face the band bending is negative, and it is positive near the Z^- face. The effect of the band bending dramatically changes the work function of the polar Z faces.[14] At the same time there is no work function difference for the antiparallel domains in the nonpolar (Y) direction. This enables us to suggest the following: *for periodically poled KTP crystals (and probably for other ferroelectric crystals with well-defined domain structures) the amplitude contrast in contact mode is due to the difference of the work functions between the opposite ferroelectric domains*.

This is supported by the calculations described in the following. Our theoretical analysis is based on a combination of two electrostatic forces acting between the tip and the ferroelectric sample. Figure 4 shows a schematic of the model used in the calculations. For a voltage V applied between the probing tip and the bottom electrode, a charge equal to $q = C(V + V_{\mathrm{cpd}})$ is induced on the tip, where C is the tip–sample capacitance and V_{cpd} is the contact potential difference between the tip and the sample. As in Ref. 3, the tip is modeled by a sphere and is replaced by an equivalent point charge that is located in its center. Due to the small Debye length in KTP crystals (~100 Å)[14] we can consider the sample as conducting and use the following expression for the tip–sample capacitance:[15]

$$C = 4\pi\epsilon_0 R + \frac{2\pi\epsilon_0 R^2}{z}, \tag{1}$$

where R is the tip radius of curvature, z is the distance between the center of the sphere and the sample surface, and ϵ_0 dielectric permittivity of vacuum.

The force on the tip includes two main interactions. The first is the Coulomb force acting between the charged tip and its image in the sample, and the second force is due to the bound polarization and free screening charges.

Using the expression for the image charge generated in anisotropic medium by the point charge situated at distance z from the boundary (see Fig. 4),[16] we calculate the force of Coulomb interaction of the charged tip with its image, induced in the sample. The component of this force at a frequency ω is

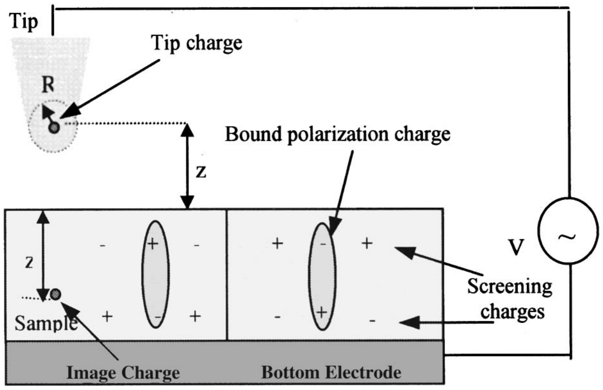

FIG. 4. Schematic of the sample and tip system used in the theoretical analysis.

$$F_{\mathrm{im},\omega}=-\frac{1}{8\pi\epsilon_0}\frac{1}{z^2}\frac{\sqrt{\epsilon_a\epsilon_c}-1}{\sqrt{\epsilon_a\epsilon_c}+1}C^2V_{\mathrm{ac}}V_{\mathrm{cpd}}, \tag{2}$$

where ϵ_a, ϵ_c are dielectric constants in the nonpolar and polar directions, respectively. The image force difference for two adjacent domains due to their asymmetric band bending can be calculated in the following way:

$$\Delta F_{\mathrm{im},\omega}=\frac{1}{4\pi\epsilon_0}\frac{1}{z^2}\frac{\sqrt{\epsilon_a\epsilon_c}-1}{\sqrt{\epsilon_a\epsilon_c}+1}C^2V_{\mathrm{ac}}\Delta\chi, \tag{3}$$

where we have used the work function of the ferroelectric in the form[17] $A_+=E_g+\chi-\Delta\chi$ for the c^+ surface of the ferroelectric, and $A_-=E_g+\chi+\Delta\chi$ for the c^- surface, with E_g the band gap of the crystal, χ the electron affinity, and $\Delta\chi$ the surface band bending.

We now describe tip–sample interaction due to the polarization and screening charges. In our model the bound polarization charges are located on the crystal surface; the screening charge is distributed in the bulk with a density $\rho(\xi)$ that is derived from Poisson equation and satisfies the equilibrium condition:

$$\int_0^\infty \rho(\xi)d\xi=-P_0. \tag{4}$$

Here P_0 is the value of spontaneous polarization of the crystal.

Taking into account the influence of the periodic polarization charge, the total electrostatic field near the ferroelectric surface can be calculated as[18]

$$E(z,x)=\frac{1}{\pi\epsilon_0}\frac{4}{1+\sqrt{\epsilon_a\epsilon_c}}\sum_{n=0}^{\infty}e^{-(2n+1)(z\pi/D)}\frac{\sin(2n+1)\dfrac{\pi x}{D}}{(2n+1)} \times\left(P_0+\int_0^\infty \rho(\xi)e^{-(2n+1)(\xi\pi/D)}d\xi\right), \tag{5}$$

where x is a coordinate in the nonpolar direction, and D is a half of the period of the domain structure. This field induces the electrostatic force acting on the AFM tip, given by

$$F_{\mathrm{el}}(z,x)=qE(z,x). \tag{6}$$

The total force difference at the frequency ω for central points of two opposite domains ($x=D/2$) is

$$\Delta F(z)=\frac{1}{4\pi\epsilon_0}\frac{1}{z^2}\frac{\sqrt{\epsilon_a\epsilon_c}-1}{\sqrt{\epsilon_a\epsilon_c}+1}C^2V_{\mathrm{ac}}\Delta\chi+2CV_{\mathrm{ac}}E(z). \tag{7}$$

This difference in the absolute value of the force between the antiparallel domains causes the domain contrast in the amplitude image.

From the EFM signal across the antiparallel ferroelectric domains, taking into account all the parameters of the experimental setup as, for example, lock-in amplifier sensitivity and spring constant of the cantilever, a force difference between two adjacent domains is calculated to be ~0.065 nN. Using a value of $2\Delta\chi\approx 40$ mV,[14] it is found that for a tip–sample distance ($z-R$) of 1 nm (which is typical for contact AFM measurements)[6] the force contrast [ΔF in Eq. (7)] is ≈0.062 nN; this is in excellent agreement with the measured value.

In summary, studies of well-defined periodic domain structure on polar and nonpolar face of ferroelectric KTP crystal enabled us to distinguish between the electrostatic and piezoelectric factors affecting the domain contrast. It is shown that the amplitude contrast in the contact regime is due to the difference of the work functions of antiparallel ferroelectric domains.

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