Ferroelectric Domain Imaging Mechanism in High-Vacuum Scanning Force Microscopy *

ZENG Hua-Rong(曾华荣)**, YU Han-Feng(余寒峰), CHU Rui-Qing(初瑞清), LI Guo-Rong(李国荣), YIN Qing-Rui(殷庆瑞)

State Key Laboratory of High Performance Ceramics and Superfine Microstructure, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 200050

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High-vacuum scanning force microscopy of the domain structures in PMN-PT single crystals is investigated. It has been shown that under high vacuum conditions, the polarization charges are not effectively compensated for by intrinsic screening charges from the ferroelectrics. This result suggests that the electrostatic tip–sample interaction plays a great contribution to the domain imaging mechanism in PMN-PT ferroelectric single crystals under high vacuum conditions.

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Scanning force microscopy in piezoresponse mode, i.e. piezoresponse force microscopy (PFM) based on an atomic force microscope, has been developed as a powerful tool for imaging and controlling of nanoscale domain structures in ferroelectric materials.[1−7] In PFM, it is assumed that the tip detects the piezoelectric vibration signal of the ferroelectric sample caused by an ac voltage applied between the tip and the counter electrode of the sample due to the converse piezoelectric effect.[1,2] The amplitude and phase of the piezoresponse signal are related to the magnitude of the piezoelectric coefficient and the direction of local polarization, respectively. However, the domain imaging mechanism in PFM depends on the tip–surface junction and imaging condition including tip radius, indentation force, and cantilever stiffness, which combine quantitatively different contributions of electrostatic and electromechanical interactions to the PFM signal,[8−14] thus causing the domain contrast mechanism to be more complex in PFM. If one of the effects prevailed over the others in certain cases, the contribution of this effect could be directly estimated and identified. Until now, most studies ascribe the domain contrast mechanism to electromechanical interactions in PFM at ambient environment, and there has been little consideration of the effect of the experimental environment such as vacuum conditions on the piezoresponse contrasts, which should be very important to understand the quantitative interpretation of the nanoscale domain (such as local ferroelectric and piezoelectric hysteresis) when SPM works in high vacuum conditions. In the present study, we performed domain imaging in $Pb(Mg_{1/3}Nb_{2/3})O_3$-30%$PbTiO_3$ (PMN-30%PT) relaxor-type ferroelectric single crystals by high vacuum scanning force microscopy in piezoresponse mode. The obtained results, which show abnormal piezoresponse behaviour related to the polarization charge compensation in PMN-PT single crystal under high vacuum environment, lead us to conclude that in high-vacuum environment, the electrostatic effect plays a dominant role in the ferroelectric domain imaging mechanism in ferroelectrics via piezoresponse mode SFM.

The piezoresponse images under high vacuum were obtained via the SFM in piezoresponse modes using a commercially available SFM instrument (SPA-300 HV, Seiko Inc, Japan). An ac voltage $V_{ac} = V_0 \sin(\omega t)$ is applied between the tip and the counter electrode of the sample during the imaging process, which gives rise to the piezoelectric vibration of the ferroelectric sample. The amplitude and phase of the piezoresponse signal are related to the magnitude of the piezoelectric coefficient and the direction of local polarization, respectively. Nanoscale field-induced displacement behaviour is measured by the detection based on the inverse piezoelectric effect. In the SFM contact mode, the conductive tip detected the piezoelectric vibration of the sample surface due to the inverse piezoelectric effect when an alternating field was applied between the tip and the sample surface. The local surface displacement information was obtained by keeping the distance between the tip and the sample constant in the z direction through the SFM feedback loop. In this experiment, all scans were performed at room temperature using a conductive Rh-coated Si cantilever (Olympus Opt. Inc.) with a spring constant of $20\,Nm^{-1}$. The vacuum condition was controlled by vacuum valve. The ac voltage for imaging domain structure has amplitude of 2 V (peak to peak) and a frequency of 9 kHz. The sample thickness is 0.3 mm.

The domain structure in PMN-30%PT single crystals were firstly visualized by the piezoresponse mode SFM at ambient environment, as shown in Fig. 1(a). The piezoresponse image in Fig. 1(a) exhibits pronounced bright and dark contrasts with narrow strip-

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** Corresponding author. E-mail: huarongzeng@mail.sic.ac.cn
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shaped ferroelectric domain patterns. The domain patterns can be interpreted as ferroelectric domains with antiparallel polarizations.[15] It is noted that the domain regions are not uniform but a mixture of small domain regions with opposite contrasts, which is similar to those observed in PZN-PT single crystals.[16,17] Figure 1(b) shows the domain patterns under 10^{-6} Torr of high vacuum (HV) from the same scanning areas as Fig. 1(a), exhibiting a domain contrast much lower than that shown in Fig. 1(a) at ambient environment. However, comparing the domain patterns in Fig. 1(a) and Fig. 1(b), it can be clearly seen that there is not a change of domain patterns but of domain contrasts. To exhibit these general phenomena under high vacuum environment, Fig. 2 shows the piezoresponse image of another scanning area in the same sample at ambient environment of 10^{-6} Torr. The domain contrasts follow the same trend as that in Fig. 1. To understand the dependence of the abnormal piezoresponse behaviour on the vacuum degree in PMN-PT single crystals, we must take into account the PFM signal due to the electromechanical response of the surface and the long range electrostatic force between the cantilever-tip assembly and the sample surface.

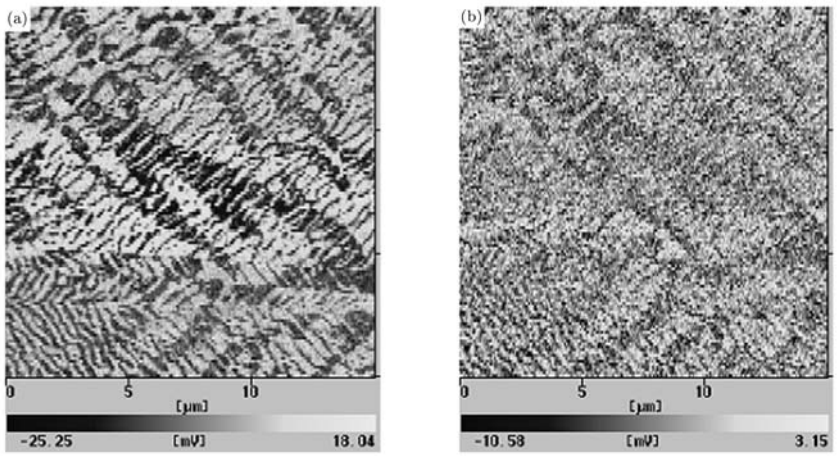


Fig. 1. Piezoresponse image of ferroelectric domains in the $15 \times 15\,\mu m^2$ scanning area of a PMN-PT single crystal (a) at ambient environment and (b) at 10^{-6} Torr high-vacuum level.

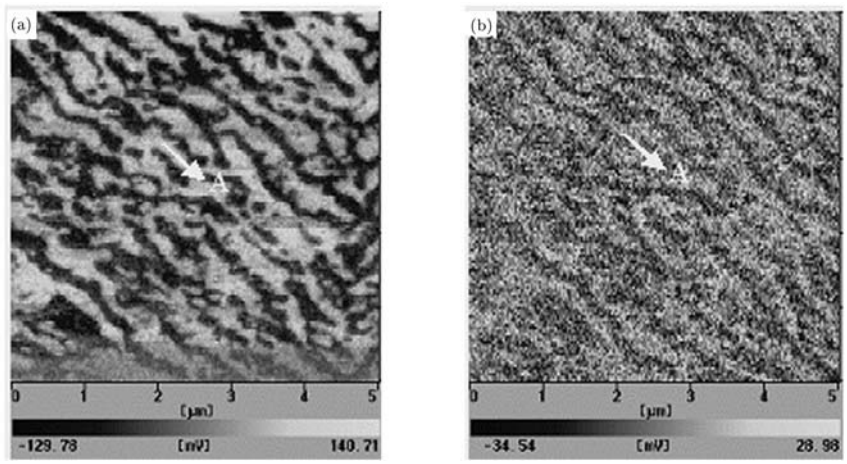


Fig. 2. Piezoresponse image of ferroelectric domains in the $5 \times 5\,\mu m^2$ scanning area of PMN-PT single crystal (a) at ambient environment and (b) at 10^{-6} Torr high-vacuum level.

Typically, the ferroelectric domains are associated with surface potential variations, i.e. surface charge distribution. In high vacuum environment, the polarization screening charges are mainly from the intrinsic electrons and holes in the ferroelectrics.[18] The presence of the intrinsic electrons and holes are insufficient for the compensation of the dipole charges, which causes the variation of surface potential on the PMN-PT surface. According to Kalinin and Bonnell,[8] the electric field above a partially screened ferroelectric surface can be expressed as $E = (1-\alpha)P\varepsilon_0^{-1}(1+\sqrt{\kappa_x\kappa_z})^{-1}$, where α is the degree of screening, P is spontaneous polarization, and κ_x and κ_z are the principal values of the dielectric constant tensor along the x and z axis, respectively. The electric field E will lead to Coulombic interaction $F_{\rm coul}$ between the surface polarization charges and SFM tip, i.e. $F_{\rm coul} = C_{\rm lol}(V_{\rm tip} - V_{\rm loc})E$, where $C_{\rm loc}$ is the SFM

tip–sample surface capacitance, $V_{\rm tip}$ SFM tip potential, $V_{\rm loc}$ is the potential of local domain directly below the tip. At the same time, the average domain size is usually much smaller than the cantilever size in the measured PMN-PT sample, thus the average surface potential on the surface of PMN-PT sample dominates non-local electrostatic contribution to PFM signal due to the buckling oscillations of the cantilever induced by capacitive cantilever-surface interactions, i.e. $F^{nl}_{\rm cap} = \frac{1}{2}C'_{nl}(V_{\rm tip} - V_s)^2$, where V_s is the surface potential averaged over the distance comparable to the cantilever length, and C'_{nl} is the non-local part due to the cantilever. In addition, the domain-related local potential directly below the tip also causes another capacitive electrostatic force, $F^{loc}_{\rm cap} = \frac{1}{2}C'_{\rm loc}(V_{\rm tip} - V_{\rm loc})^2$ between the tip and the sample surface, where $C'_{\rm loc}$ is the local part of tip–surface capacitance gradient. The increase of the Coulombic and non-local electrostatic interaction between the SFM cantilever-tip and the sample surface in high vacuum conditions would enhance the indentation force of tip to the local contact area of the sample surface. It was found in Zavala, Abplanalp and Kholkin's studies that large tip load force would severely inhibit the piezoresponse signal and reorient the polarization direction,[19–21] which results in poor domain contrasts in piezoresponse image, as illustrated in Figs. 1(b) and 2(b).

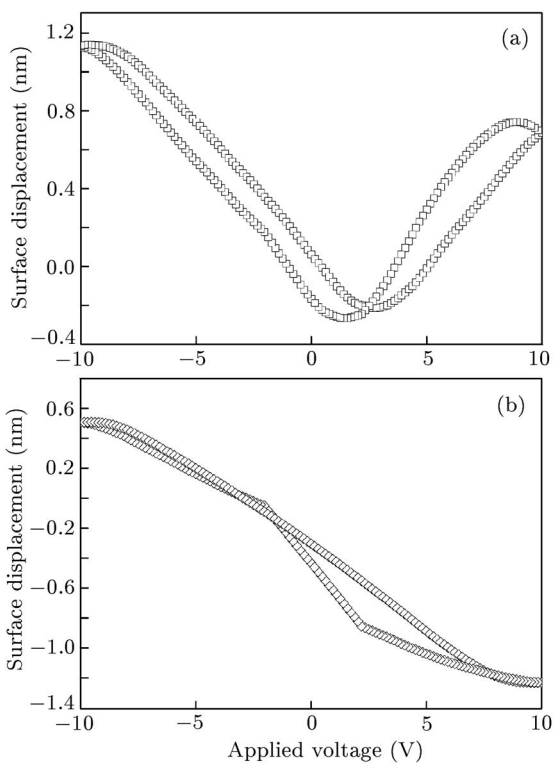


Fig. 3. Piezoresponse signal dependence of point A in Fig. 2 on the tip bias for the PMN-PT single crystal (a) at ambient environment and (b) at 10^{-6} Torr high-vacuum level.

In contrast, the Coulombic contribution can be excluded from the electrostatic tip–surface interaction in the case of ambient environment since the surface screening charges from surface adsorption almost completely screen the polarization charge on the surface of PMN-PT single crystal in air, typically $1 - \alpha \ll 10^{-3}$. Also the non-local contribution to PFM signal is very low. As a result the piezoelectric interaction prevails in the contribution to domain contrasts over electrostatic interaction in air.

As discussed earlier, strong electrostatic interaction would result in large surface indentation of PMN-PT single crystal in high vacuum conditions. The surface deformation would exhibit linear behaviour of voltage under electrostatic interaction.[8] Figures 3(a) and 3(b) give the surface deformation dependence of voltage of point A at ambient and high vacuum environment, respectively. As expected, point A exhibits a response linear in voltage due to the dominance of capacitive cantilever-surface force and cantilever buckling in the case of high vacuum condition (Fig. 3(b)), and a butterfly loop response with voltage due to the prevalence of piezoelectric interaction in air (Fig. 3(a)).

In conclusion, we have investigated high-vacuum scanning force microscopy of the domain structures in PMN-PT single crystals. It has been shown that in high vacuum environment, the polarization charges are not effectively compensated by intrinsic screening charges in the ferroelectrics. This result suggests that the electrostatic tip–sample interaction plays a great contribution to the domain imaging mechanism in PMN-PT ferroelectric single crystals under high vacuum conditions.

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