Surface potential of ferroelectric domain investigated by Kelvin force microscopy

Yunseok Kim · Seungbum Hong · Seung-Hyun Kim · Kwangsoo No

Abstract We have investigated the surface potential of poled area by varying the poled size and the sign of applied voltage on 100 nm thick $Pb(Zr_{0.25}Ti_{0.75})O_3$ films grown by chemical solution deposition using Kelvin force microscopy (KFM). In the negative poled area, as the poled size increases from 300 to 4800 nm, the domain size and the KFM contrast increased in a linear way. However, in the positive poled area, the KFM contrast increased at first and then didn't increase because of Coulomb repulsion. In two opposite poled areas, the values of the KFM contrast differed because of the internal field near the ferroelectric/electrode interface. These results imply that the surface overcharge of poled area in ferroelectric materials should be increased and the ferroelectric/electrode interface should be improved for the ultra high-density memory device.

Y. Kim (✉) · K. No
Electronic and Optical Materials Laboratory, Department of
Materials Science and Engineering, Korea Advanced Institute of
Science and Technology, 373-1, Guseong-dong, Yuseong-gu,
Daejeon, 305-701, Republic of Korea
e-mail: kseokyun@kaist.ac.kr

S. Hong
Nano Devices Laboratory, Samsung Advanced Institute of
Technology, San 14-1, Nongseo-ri, Giheung-eup, Yongin-si,
Gyeonggi-do, Republic of Korea

S.-H. Kim
Inostek Inc., 356-1, Gyeonggi Technopark, 1271-11, Sa1-dong,
Sangnok-gu, Ansan-si, Gyeonggi-do, 425-791, Republic of Korea

1 Introduction

Ferroelectric lead zirconate titanate (PZT) is one of most promising ferroelectric materials for non-volatile memory devices because of their high dielectric constant and high remnant polarization. Recently, ferroelectric domain imaging assisted by atomic force microscopy (AFM) is extensively used to study ferroelectric domain dynamics [1–3]. Among these techniques, ferroelectric domain imaging using surface potential, often called Kelvin force microscopy (KFM), is used as a nondestructive way to image ferroelectric domain at the nanometer scale [2, 3].

Domain formation in ferroelectric films has been of particular interest in non-volatile applications, such as ferroelectric random access memory (FRAM) and probe-based data storage (PBDS) [1, 2, 4]. X. Q. Chen et al. reported that the surface potential of ferroelectric domain is closely related to the pulse voltage and duration and they have used the scanning Maxwell stress microscopy (SMM) to investigate the surface potential [5]. A. Gruverman et al. investigated the asymmetric switching of ferroelectric domain in a different sign of applied voltage for the poling the ferroelectric thin films by scanning force microscopy [6]. However, they have not provided the surface potential of ferroelectric domains dependence on the poled area in a different sign of applied voltage.

In this study, we present the surface potential of various poled area in a different sign of applied voltage by KFM.

2 Experimental procedure

The sample was a 100 nm thick preferentially [111] oriented $Pb(Zr_{0.25}Ti_{0.75})O_3$ thin film on a Pt/Ti/SiO$_2$/Si substrate prepared by the sol-gel process. No pyrochlore phases were
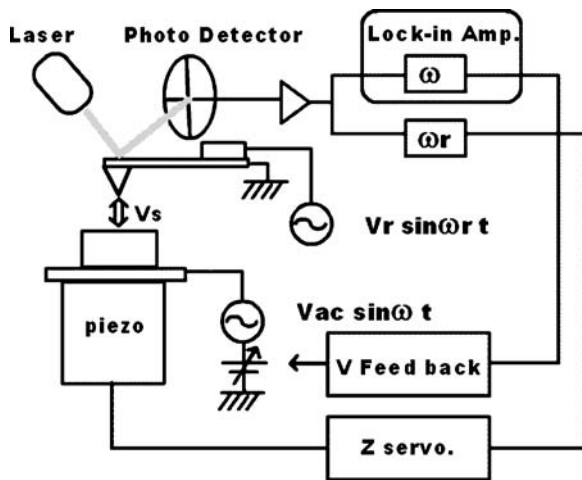
Fig. 1 Schematic diagram of the KFM equipment

detected within the detection limit of the X-ray diffraction method.

　　Initially, a writing step was performed by various scanning area with negative and positive bias voltages applied to the sample. This writing procedure was carried out in order to initialize the desired area with certain polarization directions, namely, downward and upward. The magnitudes of the bias voltages used here for polarization writing are −10 V and +10 V, respectively. The poled areas are 300 nm × 300 nm, 600 nm × 600 nm, 1200 nm × 1200 nm, 2400 nm × 2400 nm, and 4800 nm × 4800 nm, respectively. To remove the poling time effect during writing procedure, various areas of domains were formed in a same poling time/area, because the poling time (or pulse duration) affects the domain growth [7]. Then, a reading procedure was carried out by scanning 15 μm × 15 μm area with an ac signal of 7 V at 22.5 kHz. The polarization writing procedure was performed in the contact AFM mode, whereas the reading procedure was performed in the non-contact KFM mode. Figure 1 shows a schematic diagram of the KFM setup [8]. To measure the local surface charge, we simultaneously measured the surface potential and topography using a commercial AFM (Seiko, SPA400) with a gold coated Si tip (force constant of 3 N/m and reso-

nance frequency of 26 kHz). The domain size was measured from the surface potential images by taking the full width half maximum value of each centerline profile.

3 Results and discussion

　Figure 2(a) and (b) show KFM images with negative and positive poled area of 600 nm × 600 nm. In each figure, the bright regions represent the high potential and the dark regions represent the low potential. In the Fig. 2(a), the bright poled area is generated by applying the negative voltage to the sample and, therefore, the downward polarization is generated. In the downward polarization, negative bound charges are located near the sample surface. However, the KFM image reveals that the surface potential of the poled areas has an opposite contrast as compared to the oriented dipoles. This phenomenon can be explained by screen effect during the writing procedure. When an electric field is applied to the ferroelectric film by the conductive tip during the writing procedure, the charge injection from the conductive tip can occur. This injected charge compensates the bound charge of the oriented dipole. At that time, the surface adsorption, the intrinsic surface states, or the free charge in the films can also screen the bound charge [9]. The surface potentials, which is detected from KFM, of a poled area is the sum of the surface charge and the bound charge of dipoles in the poled area, as shown in the Fig. 2(c) [5]. If the amount of the surface charge is more than that of bound charge, the opposite contrast as compared to the oriented contrast can be observed. Therefore, these results of Fig. 2(a) and (b) indicate that the amount of the surface charge is greater than that of the bound charge of the remnant polarization. That is, surface overcharge is detected as a surface potential.

　　Figure 3 shows the domain size and the KFM contrast as a function of poled size. The KFM contrast indicates the potential difference between low potential and high potential in the KFM image. The poled area were varied from 300 nm × 300 nm to 4800 nm × 4800 nm. The poled area has a strong effect on the surface potential. In both the negative and the positive poled area, the domain size increases rapidly as the
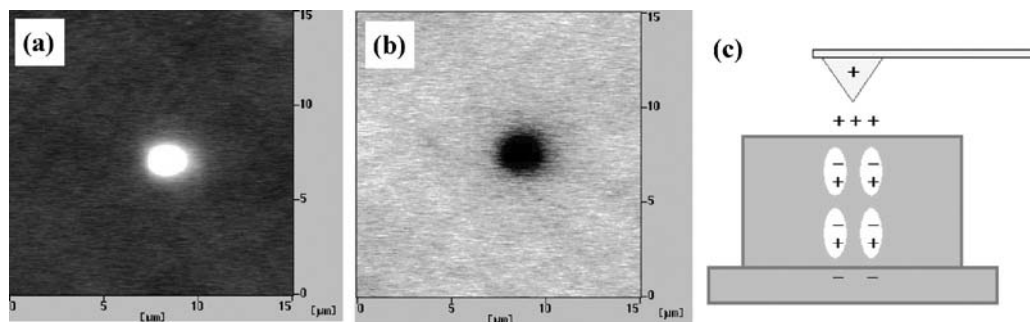

Fig. 2 KFM images with (a) the negative and (b) the positive poled area of 600 nm × 600 nm. (c) Schematic diagram in local poling of ferroelectric films
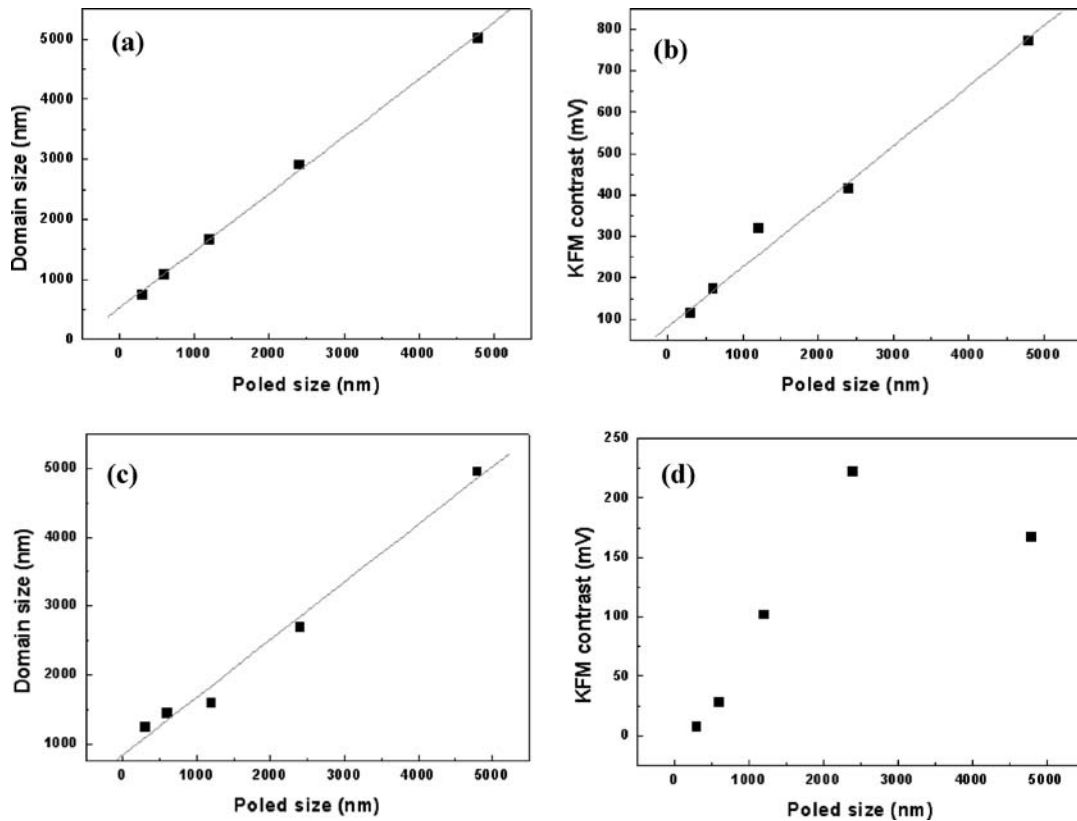
Fig. 3 Domain size as a function of poled size by applying (a) the negative voltage and (c) the positive voltage to sample. KFM contrast as a function of the poled size by applying (b) the negative voltage and (d) the positive voltage to sample

poled size increases. However, the dependence of the KFM contrast on the poled size shows different results in the negative and the positive poled area. In the negative poled area, the KFM contrast is linearly proportional to the poled size, because the amount of surface charge increases as the poled area increases. However, in the positive poled area, the KFM contrast increases rapidly at first and then doesn't increase. This phenomenon is originated from Coulomb repulsion within a surface charge [5]. This result indicates that, in the ferroelectric films, the large poled area doesn't mean the large surface potential.

On the other hand, the values of the KFM contrast are very different in two cases. The KFM contrast of the negative poled area is much higher than that of the positive poled area. This asymmetric switching property may result from the

presence of an internal built-in electric field in the PZT thin films. A built-in field could be related to the Schottky barrier between a ferroelectric with semiconductor properties and a metal bottom electrode [6, 10]. This internal field, which exists in the vicinity of the ferroelectric/electrode interfaces, doesn't depend on the applied voltage and is always pointing in one direction. This internal field is sufficiently high to polarize a bottom part of the PZT thin films, as shown in Fig. 4. In the negative poled area, the applied field is parallel to the internal field and a complete $180°$ switching can be generated. However, in the positive poled area, the applied field is anti-parallel to the internal field, as shown in Fig. 4(a), and an incomplete $180°$ switching can be generated, as shown in Fig. 4(b). From this reason, the KFM contrasts differ in two cases.

Fig. 4 Schematic diagram illustrating (a) complete $180°$ switching under a negative voltage and (b) incomplete $180°$ switching under a positive voltage applied to the sample due to the presence of the built-in internal field near the ferroelectric/electrode interface
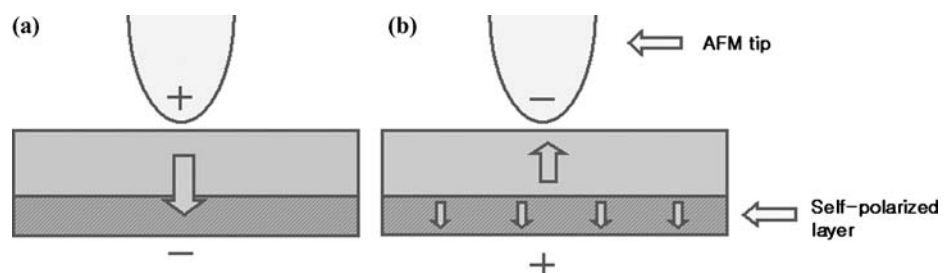
From these results, if the surface charge is used to detect the bit signal in the memory device such as resistive probe storage [4], the surface potential needs to increase sufficiently to detect the small sized domain because the domain size should be decreased for the ultra high-density. Therefore, in order to use the ferroelectric materials in the memory device, the surface overcharge of poled area should be increased. On the other hand, asymmetric switching causes a signal loss because, in the positive poled area, the internal field can not be switched and controlled. Therefore, to decrease the internal field, the ferroelectric/electrode interface should be improved.

4 Conclusion

In summary, we found that the KFM could detect a surface overcharge of poled area. In the KFM image, the surface potential of the poled areas had an opposite contrast as compared to the oriented dipoles, and an asymmetric switching in ferroelectric domains could be observed. In the negative poled area, the KFM contrast and the domain size were linearly proportional to the poled size. However, in the positive poled area, the KFM contrast increased at first and then didn't increase because Coulomb repulsion within the surface charge increases as the poled area increases. In two cases, the values of the KFM contrast were different because there is an internal field near the ferroelectric/electrode interface. The obtained result implies that the surface overcharge of poled area should be increased to decrease the domain size and the ferroelectric/electrode interface should be improved for the ultra high-density memory device.

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