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Scaling effect on statistical behavior of switching parameters of ferroelectric capacitors

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Scanning force microscopy (SFM) has been used to study nanoscale variations in switching parameters in layered perovskite films of $SrBi_2Ta_2O_9$ and to investigate the effect of capacitor scaling on the standard deviation of the capacitors' integral polarization signal. Ferroelectric poling and SFM piezoresponse imaging were performed in a number of regions on the film surface sized 2×2, 1×1, 0.5×0.5, and $0.3\times0.3\,\mu\text{m}^2$ with subsequent statistical analysis of the obtained data. It has been found that variations of the polarization signal can be approximated by the normal distribution function. The standard deviation increases with the decrease in the capacitor size, suggesting a stronger effect of grain misalignment in smaller capacitors. The obtained results imply that reliable high-density ferroelectric memories cannot be realized unless a certain capacitor size/grain size ratio is maintained. © *1999 American Institute of Physics.* [S0003-6951(99)02236-6]

Significant progress made recently in processing ferroelectric thin films and in their integration with Si-based technology has brought closer the implementation of a new generation of memory devices: nonvolatile ferroelectric random access memories.[1] The fundamental problem in developing high-density ferroelectric memories is the ability to scale capacitors to the submicrometer range and to maintain uniformity of their properties. The scaling problem was recently addressed by reports on formation and testing of nanoscale ferroelectric capacitors using layered perovskite films.[2,3] Electrical characterization of $0.7\times0.7\,\mu\text{m}^2$ capacitor arrays fabricated by means of conventional submicron lithography (Amanuma, Kunio, and Cuchiaro[2]) and self-assembled arrays of $0.2\times0.2\,\mu\text{m}^2$ capacitors (Alexe *et al.*[3]) showed reasonably good hysteresis loops. However, since a number of nanocapacitors had contributed to the measured polarization signals, the switching performance of each individual capacitor remained unchecked. Meanwhile, application of piezoresponse scanning force microscopy to the electrical characterization of individual nanocapacitors revealed remarkable variations in their switching parameters,[4] apparently due to the inhomogeneity of polycrystalline thin films at the nanoscale level. This effect may have a profound impact on the functionality of high-density memory devices: the variation of film properties at nanoscale could cause a reduced signal difference between the two logic states of a ferroelectric nanocapacitor which would lead to its failure as a memory element. Therefore, it is critically important to assess, on a submicrometer level, the effect of film crystallinity on the switching behavior of ferroelectric films. An approach, ideally suited to this purpose, is direct study of electrically induced domain transformations by means of scanning force microscopy (SFM), which offers nondestructive high-resolution imaging of domain patterns in ferroelectric thin films. Principles of piezoresponse SFM and recent advances in its application to nanoscale characterization of domain structures in ferroelectric thin films have been highlighted elsewhere.[5–8] In this study, the homogeneity of polarization reversal in $SrBi_2Ta_2O_9$ (SBT) thin films was examined at nanoscale by means of piezoresponse SFM in order to assess the scaling effect on variation in switching parameters of submicrometer ferroelectric capacitors. It should be noted that we will consider no other effect on the polarization switching signal except that of the inhomogeneity of the films, i.e., grain misalignment and secondary phases of SBT.

The SBT films were prepared by spinning a precursor solution (Sr/Bi/Ta=0.8/2.2/2.0) on silicon wafers with a thermal 300-nm-thick SiO_2 layer and sputtered layers of Ti (25 nm) and Pt (180 nm). The spinned-on layers were heat treated at 250 °C for 7 min and then crystallized using RTA at 760 °C for 30 s. A postannealing process, which was performed for 1 h at 800 °C in O_2, produced 170-nm-thick SBT films. Well-shaped hysteresis loops were obtained using Pt top electrodes. Remanent polarization $2P_r$ and coercive field E_c were found to be 20 μC/cm^2 and 43 kV/cm, respectively, for an applied voltage of 5 V.

In the present study, a commercial force microscope PSI Autoprobe CP was used. The SFM experimental procedure for investigation of the scaling effect on variation of switching parameters was as follows. We performed ferroelectric poling (this procedure involved scanning a selected region of the film without the top electrode with the conductive SFM tip held under a dc bias) and piezoresponse imaging of a number of regions on the film surface of 2×2, 1×1, 0.5×0.5, and $0.3\times0.3\,\mu\text{m}^2$ in size (more than 30 regions of each size) with subsequent histogram analysis of the resulting piezoresponse images using the ULTIMAGE 2.5.1 image processing program. After this, statistical treatment of the obtained data was carried out. An ac voltage with an amplitude of 3.2 V and a frequency of 10 kHz was used for domain imaging.

Preliminary SFM studies of polarization reversal in SBT films at nanoscale have already shown significant variations of hysteresis loop parameters from grain to grain as well as variations of domain contrast within the poled regions.[4,9–11] It has been suggested that this effect is primarily due to ran-

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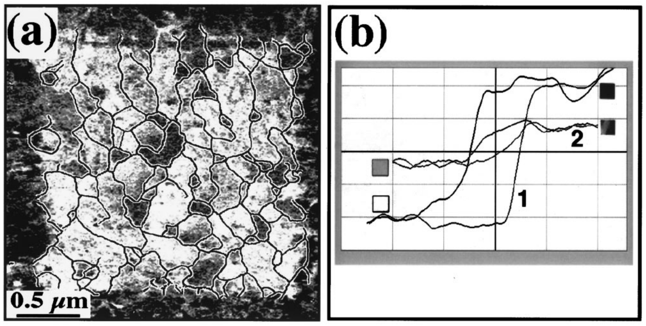

FIG. 1. Ferroelectric switching in the SBT film by means of SFM: (a) piezoresponse image showing a polarized $2\times 2\ \mu\mathrm{m}^2$ region at the center produced by scanning the tip under a dc bias. Black lines indicate positions of grain boundaries. (b) Typical hysteresis loops observed in differently oriented grains: (1) *a*-oriented grain and (2) randomly oriented grain.

domly oriented grains, although it may be also affected by the presence of secondary nonferroelectric SBT phases. Figure 1 shows a typical piezoresponse image of the SBT film after poling and nanoscale hysteresis loops obtained from differently oriented grains. Obviously, these nanoscale variations of local switching parameters, being averaged over the capacitor area, do not seriously affect the uniformity of switching parameters of relatively large capacitors ($>100\ \mu\mathrm{m}^2$). However, with the decrease in the capacitor size the specific contribution of each grain to the capacitor's polarization signal increases. While it is difficult to say how the coercive voltage of a given capacitor will be influenced by variations in the coercive voltages of different grains, it is possible to estimate the effect of grain misalignment on the capacitor's polarization signal by analyzing the distribution of the piezoresponse signal within the poled region.

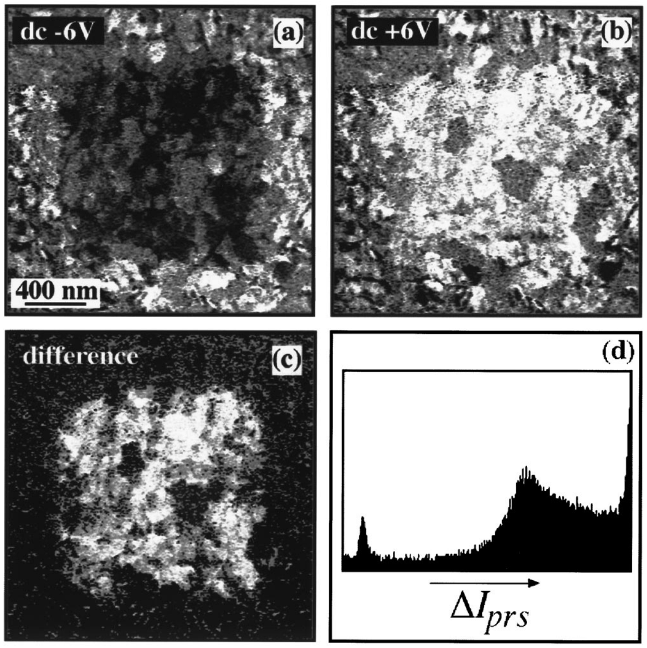

FIG. 2. Piezoresponse images of a sol-gel SBT film after its central part of $1\times 1\ \mu\mathrm{m}^2$ was poled by applying a negative (a) and positive (b) bias of 6 V; (c) differential piezoresponse image. Areas of maximum intensity are the best switchable regions, while areas of low intensity within the central bright square are barely switchable; (d) histogram of the differential piezoresponse image showing a distribution of the number of pixels per ΔI_{prs} value. The larger ΔI_{prs} value corresponds to the higher degree of switchable polarization.

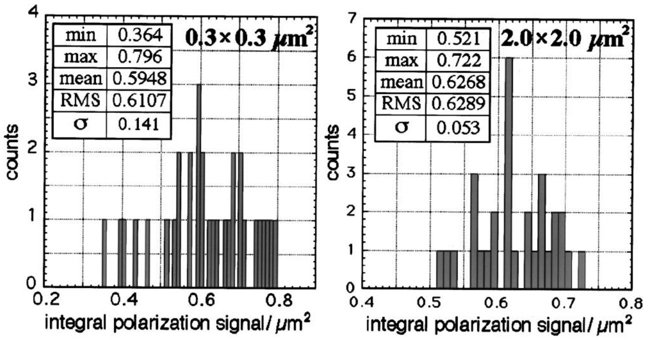

FIG. 3. Distribution of the integral polarization signal P_{int} for 0.3×0.3 and $2\times 2\ \mu\mathrm{m}^2$ capacitors. The P_{int} values are expressed as fractions of the maximum theoretical polarization signal $P_{\mathrm{int}}^{\mathrm{max}}$.

Generally, a digital piezoresponse image (in our case of 256×256 pixels) represents a two-dimensional map of the light intensity which at each point corresponds to the first harmonic signal I_{prs} (local piezoresponse signal). This signal can be expressed as $I_{\mathrm{prs}}=kA\cos\phi$, where k is a calibration constant, A is the vibration amplitude of the film and $\cos\phi$ is the phase difference between the first harmonic signal and an external modulation (imaging) voltage V applied across the film. The vibration amplitude is determined by the piezoelectric coefficient d_{33} and the imaging voltage: $A=d_{33}V$. In a monodomain ferroelectric, the piezoelectric coefficient can be expressed as a function of its polarization P

$$d_{33}=2Q_{11}\epsilon\epsilon_0 P, \tag{1}$$

where Q_{11} is the electrostriction coefficient, which only weakly depends on external parameters, and ϵ is the dielectric constant (the validity of this relation for SBT thin films was confirmed by Kholkin, Brooks, and Setter[12]). Therefore, at each point, or pixel, of the piezoresponse image the piezoresponse signal I_{prs} is proportional to the normal component of the local polarization. Finally, the integral polarization signal from a poled region, which is proportional to the remanent polarization of a capacitor of the corresponding size, can be found by summing up the piezoresponse signals I_{prs} of all the pixels comprised in this region.

To improve the method of evaluating capacitor switching performance, we suggest that the same area of the film is poled in opposite directions by applying opposite voltage biases to the probing tip with subsequent differential analysis of the resulting piezoresponse images [Figs. 2(a) and 2(b)]. During differential analysis, two images of the same area, those of positively and negatively polarized film, are compared so as to produce a differential piezoresponse image [Fig. 2(c)] where a signal at each pixel is defined as $\Delta I_{\mathrm{prs}}=I_{\mathrm{prs}}^{+}-I_{\mathrm{prs}}^{-}$, so that the best switchable regions appear with maximum intensity (bright) and the nonswitchable regions exhibit the lowest intensity (dark). Generally, the ΔI_{prs} signal at each pixel of the differential image is proportional to the degree of switchable local polarization

$$\Delta I_{\mathrm{prs}}=I_{\mathrm{prs}}^{+}-I_{\mathrm{prs}}^{-}\sim 2VQ_{11}\epsilon\epsilon_0(P^{+}-P^{-}), \tag{2}$$

where P^{+} and P^{-} are normal components of the local polarization for a positively and negatively polarized region, respectively. Histogram analysis of the differential piezore-

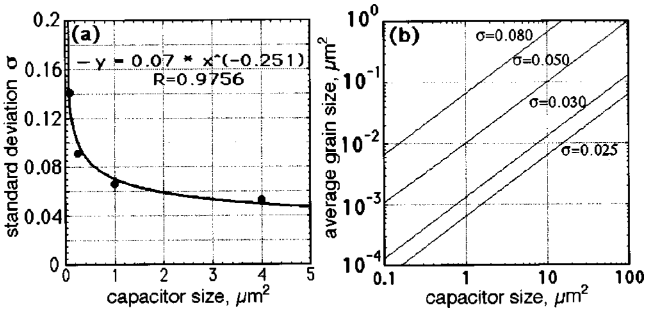

FIG. 4. (a) Capacitor size dependence of the standard deviation for the SBT film (the average grain size is about 0.04 μm^2); (b) average grain size vs capacitor size for different values of $\boldsymbol{\sigma}$.

sponse image reveals a quantitative distribution of pixels per ΔI_{prs} value [Fig. 2(d)]. By integrating the histogram data, the integral switching polarization signal P_{int} for a capacitor of the corresponding size can be estimated. It is important to note that this method allows one to exclude regions with pinned domains from the estimation of the P_{int} value as well as regions with nonferroelectric structure. The distributions of the integral polarization signals for capacitors of a specific size were analyzed using standard statistical methods. To be able to compare data for capacitors of different sizes, polarization signals for capacitors of a given size were normalized with respect to their maximum theoretical value $P_{\mathrm{int}}^{\mathrm{max}}$ per unit area. (The maximum theoretical value of P_{int} is that of a capacitor with perfectly *a*-oriented uniform SBT film).

Distributions of the integral polarization signal for 2×2, and $0.3\times0.3\ \mu$m^2 capacitors are shown in Fig. 3. It has been found that the distributions can be well approximated by the normal distribution function. A parameter of particular importance here is the standard deviation $\boldsymbol{\sigma}$, as it indicates the degree of inhomogeneity of the switching properties of the capacitors. From the insets in Fig. 3 it can be seen that the standard deviation increases with the decrease in the capacitor size: $\boldsymbol{\sigma}$ is 0.053 for $2\times2\ \mu$m^2 capacitors while it is 0.141 for $0.3\times0.3\ \mu$m^2 capacitors. The standard deviation of P_{int} against the capacitor size can be fitted by the power law dependence with the power index of -0.251 [Fig. 4(a)]. On the other hand, the mean value of the integral polarization signal is almost the same for capacitors of all sizes and is approximately $0.60P_{\mathrm{int}}^{\mathrm{max}}$.

Obviously, the standard deviation of P_{int} is strongly affected by the A_c/A_{gr} ratio (A_c-capacitor size, A_{gr}-average grain size) which, simply speaking, shows the average number of grains incorporated into a capacitor. Given that the average grain size in our SBT film sample is about 0.04 μm^2, this ratio will be 2.25 for the $0.3\times0.3\ \mu$m^2 capacitor and 100 for the $2\times2\ \mu$m^2 capacitor. Assuming that the orientation distribution of grains is not affected by a change in the average grain size, it is possible to estimate the number of grains which a capacitor of a given size should incorporate in order to maintain a certain value of the standard deviation. For example, from Fig. 4(a) it can be found that the standard deviation $\boldsymbol{\sigma}$ of 0.025[13] can be achieved only for the 62 μm^2 capacitor (A_c/A_{gr} *ratio* is about 1500). For a high-density Fe random access memory (FeRAM) of 16 Mbit the capacitor size should be about $1.0\times1.0\ \mu$m^2.[14] Therefore, to meet the requirement of $\boldsymbol{\sigma}\leqslant0.025$, all capacitors should incorporate at least 1500 grains and the average grain size in this case must not exceed $6.5\times10^{-4}\ \mu$m^2 (about 25 nm in linear size). Plots of the average grain size against the capacitor size for different values of $\boldsymbol{\sigma}$ are shown in Fig. 4(b). So, from statistical analysis of the integral polarization signal, it is apparent that, in order to provide high uniformity of switching properties, nanoscale SBT capacitors should utilize films with grains much finer than several hundred nanometers (in lateral size). The grain size can be estimated from the capacitor size/grain size ratio for an acceptable value of $\boldsymbol{\sigma}$ [Fig. 4(b)]. Although the assumption of no grain size effect on grain orientation may not seem quite realistic, it may serve as a first approximation for evaluation of the scaling effect on homogeneity of switching parameters of nanoscale capacitors.

In conclusion, using the piezoresponse SFM imaging method, the scaling effect on variation of integral polarization signal from nanoscale $SrBi_2Ta_2O_9$ capacitors has been analyzed. Differential piezoresponse imaging was suggested for improving the estimation method. It has been found that variations in the integral polarization signal for the capacitors in the range from 0.3×0.3 to $2\times2\ \mu$m^2 in size can be approximated by the normal distribution function. The standard deviation of the signal increases with a decrease in the capacitor size following a power law dependence. It is suggested that for implementation of high-density nonvolatile ferroelectric memories a certain capacitor size/grain size ratio should be maintained to provide high uniformity of switching properties of nanoscale capacitors. Another option would be to use epitaxial monocrystalline films.

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