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Nanoscale Investigation of a Ferroelectric Domain and Its Electric Conduction Mechanism

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We report on the nondestructive investigation and visualization of ferroelectric film using scanning probe microscopy (SPM). By using an Au coated SPM tip as a mobile top electrode, local (less than 90 nm in diameter) and large-scale ($1\times1\mu m^2$) polarized domain were formed by applying a DC voltage between the tip and bottom electrode and observed by monitoring the piezoelectric vibration of the ferroelectric film as induced by a small AC modulation voltage. Moreover, local current-voltage measurements revealed that the current through the Au-tip/PZT/$SrRuO_3$ heterostructure was rectified depending on the direction of polarization. Analysis by combining these SPM techniques provided insight into the structure, conductance, and ferroelectric properties in the nanoscale region of the ferroelectric films.

Key words; SPM(SFM), piezoelecric response, PZT, SBT

1. INTRODUCTION

Ferroelectric thin films are currently of interest in technological applications for nonvolatile memory devices because of their switchable polarization. In order to improve ferroelectric film properties further study, such as, local conductance behavior, the exact nature of the domain structure, and the domain dynamics on the nanometer scale, is necessary.

Recently SPM has been widely employed for nondestructive inspections of ferroelectric films[1-12)], including such aspects as formation and imaging of nanoscale ferroelectric domains[3-6)], studing switching behaviors[7-9)], and observations of the leakage current characteristics[10,11)] and transition[12)].

In this paper, by using the SPM conductive tip as a mobile top electrode, leakage current path and local current-voltage (I-V) characteristic through ferroelectric film were measured by contact mode atomic force microscopy (AFM). Local and large-scale polarized domains were formed by applying a DC voltage between the tip and bottom electrode and were observed by monitoring the piezoelectric vibration of the ferroelectric film as induced by a small AC modulation voltage.

2. EXPERIMENTAL

Experiments were carried out using three samples:

1) $Pb(Zr,Ti)O_3$ film epitxially grown with the laser ablation method on an $SrRuO_3/SrTiO_3$ substrate characterized by X-ray diffraction (XRD) measurements.
2) $Pb_{1-x}La_x(Zr,Ti)O_3$ film produced by the sol-gel technique on $IrO_2/Pt/SiO_2/Si$ substrates with about 90% of the film (111) textured as estimated from XRD peak intensity and FWHM.
3) $SrBi_2Ta_2O_9$ film produced by the sol-gel technique on $Pt/Ti/SiO_2/Si$ substrates.

Hereafter we will designate these as PZT, PLZT, and SBT respectively.

SPM studies of ferroelectric films were performed using a commercial SPM (Seiko Instruments SPI3800) which was operated in air. An Au (70 nm thick)/Cr (10 nm thick) coated Si cantilever was used with a spring constant and resonant frequency of 0.23 N/m and 12 kHz, respectively.

The electrical conductances of the Au-tip/ferroelectric-film/bottom-electrode can be measured simultaneously with a contact mode AFM, which has a feedback system to maintain a constant repulsive coupling force between the tip and the sample surface. The topographic images were observed by detecting the deflection of a cantilever by an optical beam deflection system while scanning the sample surface.

The current image was obtained by

conduction in a ferroelectric single grain, Watanabe et al.11) have investigated for $BaTiO_3$ film grown on n-$SrTiO_3$ by using Pt coated AFM tip as a top electrode. They compared the difference between the I-V characteristic of nanometer and millimeter-size Pt/$BaTiO_3$ contacts. The marked difference between them is that nanometer size electrode exhibited a very high resistivity and an abrupt breakdown while millimeter size electrode showed Schottky diode-like characteristic.

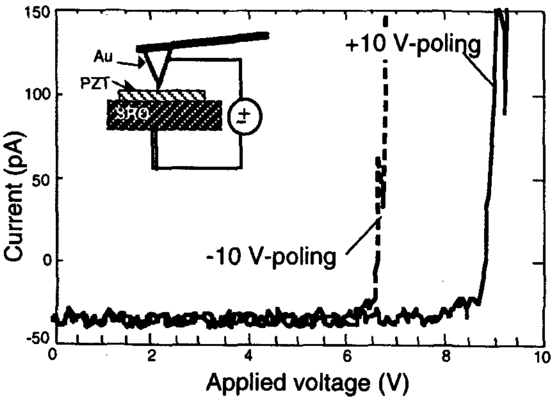

Fig.3 Poling voltage dependent I-V characteristic for Au-tip/ PZT/SRO heterostructure.

3.2 Local I-V measurement

Local I-V measurements for the epitaxialy grown PZT film were performed as a function of a DC poling voltage. After the poling voltage pulse (± 10V, 100 msec)13) was applied between the Au-tip and the SRO bottom electrode, a DC voltage was applied from 0 to 10 V to measure the reverse current through the Au-tip/PZT contact. Typical I-V characteristics for the Au-tip/±10V-poling PZT/SRO heterostructure are shown in Fig.3. According to the figure, the I-V characteristic after +10 V poling switched to the low-current phase rather than with -10 V poling. Nanometer scale I-V characteristic exhibited rectifier depending on the poling voltage.

Utilizing the rectification of I-V characteristic, a domain pattern which is written by applying a DC voltage, can be read out by detecting the change in bistable conduction properties. The rectifier conduction properties can be explained by a model of the polarization dependence of band bending of the Au-tip/PZT contact14).

3.3 Control and imaging of a local ferroelectric domain

Polarization induced patterns can be formed on the ferroelectric film by applying a DC voltage and can be observed using piezoelectric response method. We demonstrated the write operation on the epitaxialy grown PZT film.

First, a piezoelectric loop was obtained by superimposing a poling voltage (± 8 V) of a lower frequency (15 mHz) onto the small AC modulation voltage (8 kHz, 0.5 Vrms). As can be seen in Fig.4, the resulting curve shows hysteretic behavior. In this figure, the positive piezoresponse means an increase in the film thickness and corresponds to the phase with downward polarization. As-grown film showed a negative piezoresponse which corresponds to the phase with upward polarization and changes its sign after applying the positive voltage (greater than the threshold voltage) to the tip. Note that the piezoelectric response of as-grown film showed uniform negative magnitude (polarization upward); the antiparallel (polarization downward) domain can be formed by applying a positive voltage to the tip.

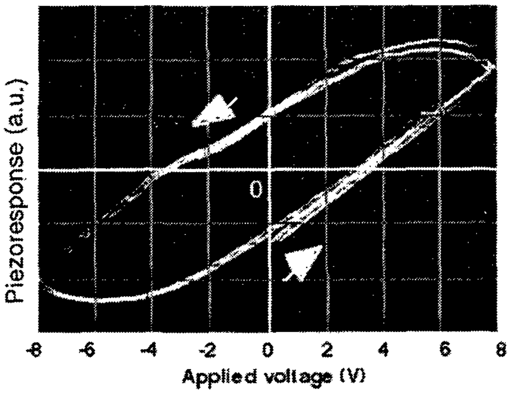

Fig.4 Piezorenponse hysteresys loop for Au-tip/PZT/SRO heterostructure.

At four positions within a 500 nm× 500 nm area, 50 msec pulse voltages of 10 V were applied to the tip. Figure 5(a) and (b) show topographic and piezoresponse images of the written area, respectively, taken simultaneously. The applied pulse voltages showed almost no effect on the topographic image, whereas the four bright dots pattern can clearly be seen in the piezoresponse image and monitoring the current while scanning the tip at a fixed bias voltage. The local I-V characteristic of a specific point was also measured by applying the bias voltage between tip and the bottom electrode. The polarized domain was formed by applying a pulse voltage between the tip and the bottom electrode.

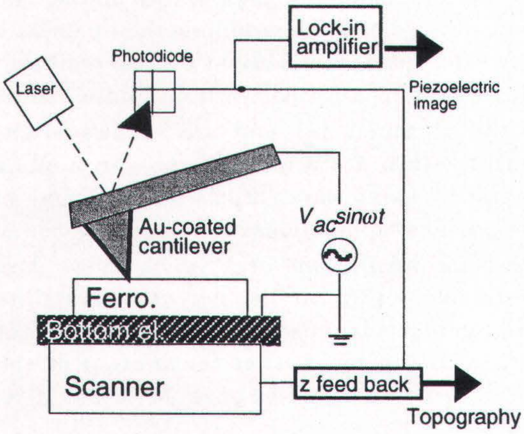

Fig.1 The schematic diagram of the SPM applied for piezoelectric measurement.

The polarized domains were observed by the SPM piezoresponse method. The piezoresponse method is based on the detection of the film vibration caused by the external ac field applied to the film through the SPM tip. A schematic diagram of the SPM piezoelectric measurement, which is similar to that recently reported by Franke et al.[1)], Hidaka et al.[3,4)], and Gruverman et al.[5,7,9)] is shown in Fig.1. An AC modulation voltage with an amplitude of 0.5-2V and a frequency of 8 kHz below the cantilever resonant frequency was applied between the tip and the bottom electrode, and the signal of the photodiode was fed into a lock-in amplifier. (The Au-tip was positively biased with respect to the bottom electrode.) The lock-in amplifier monitored the first harmonic signal which corresponds to the change in film thickness caused by the inverse piezoelectric effect. Moreover, the phase of the output signal depends on the sign of the piezoelecric coefficient which depends on the polarization direction.

3. RESULTS AND DISCUSSION

3.1 Leakage current path

The leakage current path of 45-nm thick SBT film (coated one time in the sol-gel process) was investigated. Figure 2(a) and (b) show the topographic image and current image, respectively, taken simultaneously at a voltage of 7 V applied to the tip. The dark area in Fig.2(b) corresponds the area in which the current flows. By comparing these two images, the leakage current path can be located along the grainboundaries. The film must be getting thinner at the grainbaoundary. Typical I-V curves for grainboundary (solid line) and for a single grain (dotted line) are shown in Fig.2(c). Little current were observed until the bias voltage first exceed 6.5 V (for grain) and 11 V (for the single grain) and avalanche-like breakdown were observed at higher bias voltage. For a 180-nm thick SBT film (coated four times in the sol-gel process), little current was observed, even when bias voltage was increased as high as 30 V.

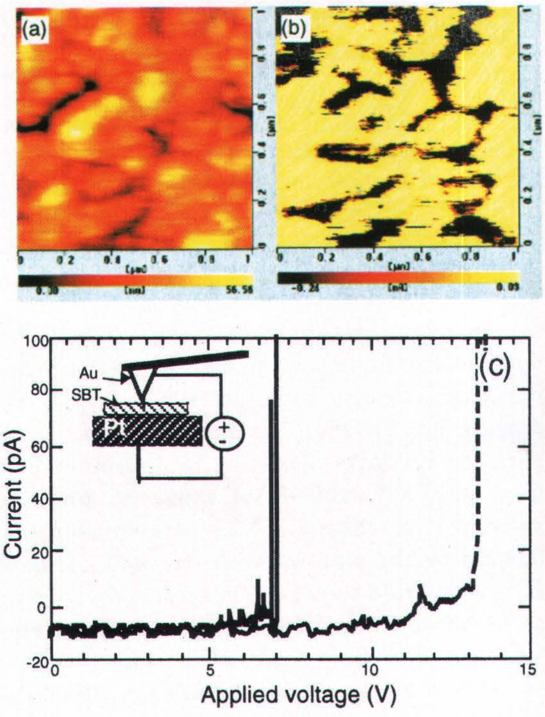

Fig.2 Leak current characteristic of the 45 nm thick SBT film: simultanelously obtained (a) topograpic and (b) current (at V_{biased} = 7 V) images and (c) typical I-V curves for grainboundary (solid line) and for a single grain (dotted line).

Fujisawa et al[10)]. reported that the current through CVD-grown PZT film also flowed through the grain boundaries. As for the exhibits the opposite piezoelectric response than the surrounding dark area. The bright dot which stands below to the left, is broken off due to a large precipitate shown in the topographic image (Fig.5(a)). According to the line profile along the dotted line shown in Fig.5(b), the size of a bright dot is about 90 nm.

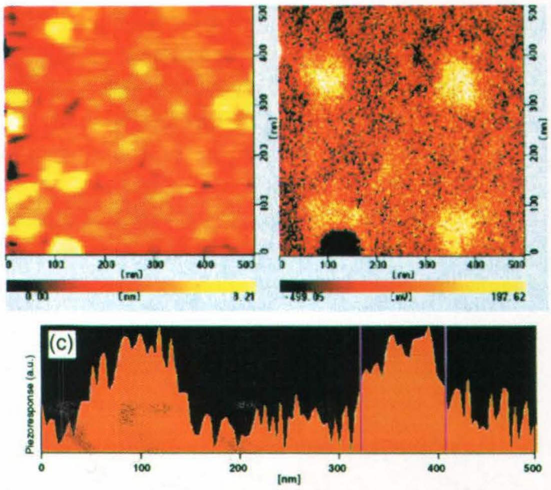

Fig.5 Simultaneouly obtained (a) Topographic and (b) piezorenponse images pf the PZT film. (c) A cross section alomg the dotted line of the piezoresponse image (b).

The limitation in recording density of ferroelectric film is not only an interest in basic research but also of a practical concern for application to high density FeRAM. We performed a write operation in a thinner PZT film with a thickness of about 35 nm and succeeded in observing the written polarized domain by the piezoresponse method. Hidaka et al.[3] reported that a polarized domain as small as 30 nm which was of the order of the tip radius, was formed in 45 nm thick PZT film.

It has been demonstrated that this SPM technique is useful for investigation of domain dynamics. Gruverman et al.[9] directly observed the retention characteristic for a single grain of PZT film and showed the spontaneous polarization reversal nucleates at the grain boundary. Colla et al. [8] showed the evolution of the frozen polarization region during the fatigue in Pt/PZT/Pt capacitor. The degree of fatigue (corresponds to the potion of the frozen polarization region) obtained by analyzing the piezoresponse image are in good agreement with those obtained by standard polarization measurements.

3.4 Large scale switch behavior

We performed local and large-scale switching in PLZT film. The topographic and piezoresponse image of the PLZT film are shown in Fig.6(a) and (b), respectively. In advance of the measurement, an area of 1×1 μm² at the center was polarized by scanning the tip under a 10 V bias. The dark and bright regions represent the area of opposite polarization phase. Most of the polarized 1×1 μm² area shows bright contrast while about a half of the surrounding as-prepared area shows bright contrast. One of the possible explanations of the small portion of the polarized 1×1 μm² area which shows dark contrast is that it is due to crystallite misalignment, because it is in close agreement with XRD the result that the content of the disordered or misoriented phase are about 10 %.

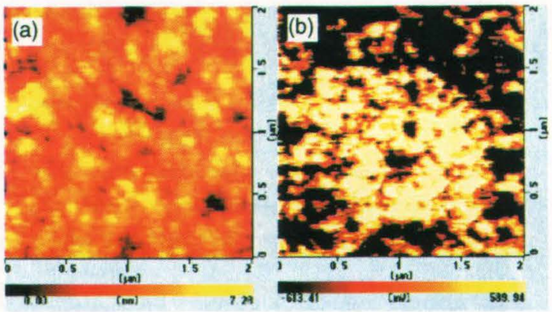

Fig.6 Simultaneouly obtained (a) Topographic and (b) piezorenponse images pf the PZT film. In advance of the measurement, 1×1 μm² area at the center were scanned by the tip under a dc poling voltage of +10 V.

4. CONCLUSION

We have reported results on a nanoscale investigation of ferroelectric film using SPM. The comparison between current image and topographic image revealed that the current path in SBT thin film was along the grainboundaries. The local I-V characteristic for Au-tip/PZT/$SrRuO_3$ heterostructure showed rectifier depending on the direction of polarization. Furthermore, the polarized domains, the 90 nm dots and 1×1μm² area were formed by applying DC voltages between the tip and bottom electrode and observed by vibration of the ferroelectric film as induced by a small AC modulation voltage. From the studies discussed here, it was found that the analysis combining these SPM techniques provided insight into the structure, conductance, and ferroelectric properties of the nanoscale region of the ferroelectric films.

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