Direct Observation of Potential Distribution across $PbZr_xTi_{1-x}O_3$ Thin Films Using Off-Axis Electron Holography

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This paper describes how we used off-axis electron holography to observe the potential distribution across a ferroelectric $PbZr_xTi_{1-x}O_3$ (PZT) capacitor and the surrounding electric field created by spontaneous polarization of polycrystalline PZT (poly-PZT) thin film. We produced equi-phase contour maps of the incident electron beam. Because the phase of the beam was shifted according to the electric field strength in the PZT thin film, these contour maps can be used to visualize the electric field. This is the first electron-holographic representation of the electric field of a poly-PZT thin film of a ferroelectric memory device.

1. Introduction

Recently, both academic and industrial interests have converged on ferroelectric thin films (e.g., $PbZr_xTi_{1-x}O_3$ [PZT] deposited by sputtering, chemical vapor deposition, or chemical solution deposition).[1)-3)] This interest is due to the importance of these films in the capacitors of ferroelectric random access memory (FeRAM).

Among the electrical properties of ferroelectric thin films, spontaneous polarization is the most important, because it strongly affects the performance and reliability of an FeRAM's retention and fatigue properties.

To characterize the polarization of a ferroelectric thin film, the hysteresis of polarization is usually measured with a large-area, electrically biased capacitor having flat and parallel electrodes. Since such a capacitor is far larger than the devices used in real applications and is usually manufactured using a much simpler process, its electrical properties do not necessarily reflect the performance of real devices. Therefore, we must characterize the ferroelectric films of real devices.

Conventional transmission electron microscopy (TEM) is naturally useful in the crystallographical study of thin films of such small devices, since the ferroelectricity of PZT film is closely related to its crystal structure. For example, TEM provides a lot of information regarding the domain structure in a poly PZT grain. Unfortunately, TEM does not allow us to visualize the electric potential in and outside the thin film that is introduced by the ferroelectric film itself, so we cannot recognize whether the film is ferroelectric. Therefore, other powerful means for studying PZT films should be developed. For this purpose, we tested a new method of characterizing the ferroelectricity of a capacitor film by means of off-axis electron holography using TEM.

In electron holography, visualizing the phase change of an electron wave affected by a local scalar and/or vector potential makes it possible to directly image the distribution of the electric or magnetic field at high resolution.[4),5)] Therefore, electron holography could be a valuable tool in the study of ferroelectric materials. This method was used in the study of bulk ferroelectric materials.[6)]

In this method, the electron phase change resulting from the polarization change across the domain wall enables a direct measurement of both the domain wall thickness and the spontaneous polarization. This method was also used in a quantitative study of space-charge distribution at the grain boundary of electroceramics.[7)]

This paper describes how we applied electron holography to observe potential distributions across a ferroelectric capacitor and the surrounding ferroelectric thin film.

2. Experiment

2.1 Principle of off-axis electron holography

Figure 1 shows the principle of off-axis electron holography.[8)] The technique uses a field emission type TEM equipped with an electron biprism that splits the incident beam into two parts. Half of the beam, the reference wave, travels outside the specimen and the other half, the scattered or diffracted wave, passes through the specimen. The two waves are combined at the other side of the specimen to form a holographic electron interference pattern. When passing through the local electric field of the specimen, the scattered wave undergoes a differential phase shift, which bends the interference fringe. The extent of this bending depends on the distribution of the electric field, making it possible to measure the field strength from the distortion in the interference pattern. Since the phase change between the fringe maximums is 2 , the fringe shift measurement permits a quantitative determination of the phase change to be made. Hence, the magnitude of the polarization vector can be found from the phase image, which reveals the electro-magnetic potential distribution in and outside the ferroelectric thin film.

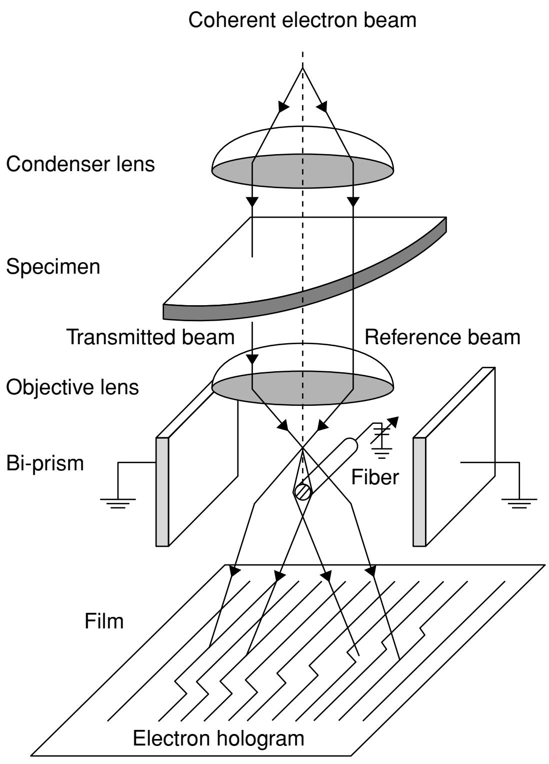

Figure 1
Principle of off-axis electron holography.

2.2 Samples and equipment

For this study, polycrystalline PZT (poly-PZT) film samples 250 to 280 nm thick were used. The films were deposited on Pt/Ti bottom electrodes by sputtering. The bottom electrodes were deposited on an SiO_2 layer on 6-inch (001) Si wafers. A Pt top electrode about 150 nm thick was also deposited. The dopant in the PZT was La. We prepared three types of samples for TEM and holography observation: 1) a PZT film with a bottom electrode, 2) a PZT film with a top and bottom electrode, and 3) the PZT capacitor of an FeRAM device. Cross sections of the samples were prepared using a consistent cutting, lapping, and final ion-etching process. For hologram observation, we used a JEOL-JEM2010FEG with a hologram unit.

3. Results and discussion

3.1 Cross section of PZT/Pt-Ti (PZT film with bottom electrode)

Figure 2 (a) shows a cross-sectional TEM image of the PZT and the bottom electrode. The bottom electrode remains adhered to one side of this sample. The bottom electrode is in physical contact with the PZT. The electrode is also connected to ground potential through the sample holder of the TEM. Both the upper side of the PZT and the bottom side of the electrode are exposed to vacuum.

Figures 2 (b) and **(c)** show a hologram of the cross section shown in Figure 2 (a) and a phase image of the capacitor obtained from the hologram, respectively.

In Figure 2 (c), in the vacuum region adjacent to the PZT film, the contour pattern indicates the change of electron phase. The contour lines correspond to an equi-electric potential line, showing the electric field distribution due to the polarization of the PZT film. In other words, the PZT film polarizes spontaneously, indicating that it is ferroelectric. There is no contour contrast in the vacuum region of the electrode side, which clearly indicates that the electric field due to the polarization of the PZT film is shielded by the residual electrode.

It is evident that this reconstructed image reflects the real potential distribution surrounding the PZT film and bottom electrode system, because the shielded area has been successfully reconstructed.

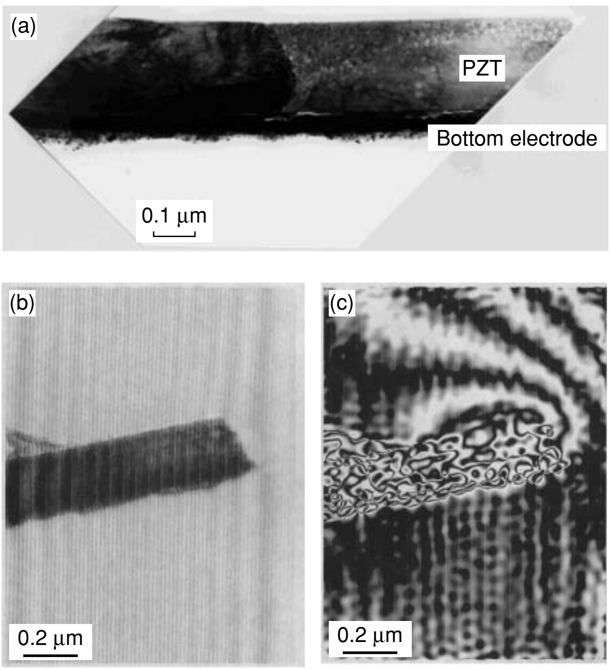

Figure 2
(a) Cross-sectional TEM image of PZT and bottom electrode. (b) Hologram of cross section shown in (a).
(c) Phase image of capacitor obtained from hologram shown in (b).

3.2 Cross section of Pt/PZT/Pt-Ti (PZT film with top and bottom electrodes)

Figures 3 (a) and **(b)** show a cross-sectional hologram of the Pt/PZT/Pt film and the corresponding reconstructed phase image, respectively. They also show the electron phase change due to the coulomb potential (S1,S2); the phase change in this figure has been magnified 5 times.

In this case, the top electrode was isolated and electrically floating, then a charge was induced and distributed on the surface (the bottom electrode was connected to ground potential). Therefore, there are contour patterns (S1) in the vacuum region surrounding the electrode, which reflect the strength distribution of the electric potential field induced by the surface charge on the electrode.

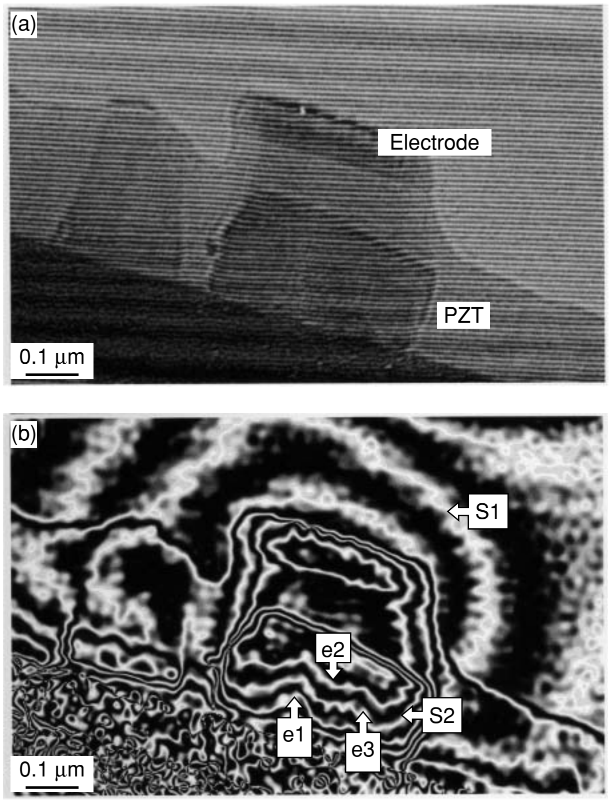

Figure 3
(a) Cross-sectional hologram of Pt/PZT/Pt. (b) Reconstructed phase image.

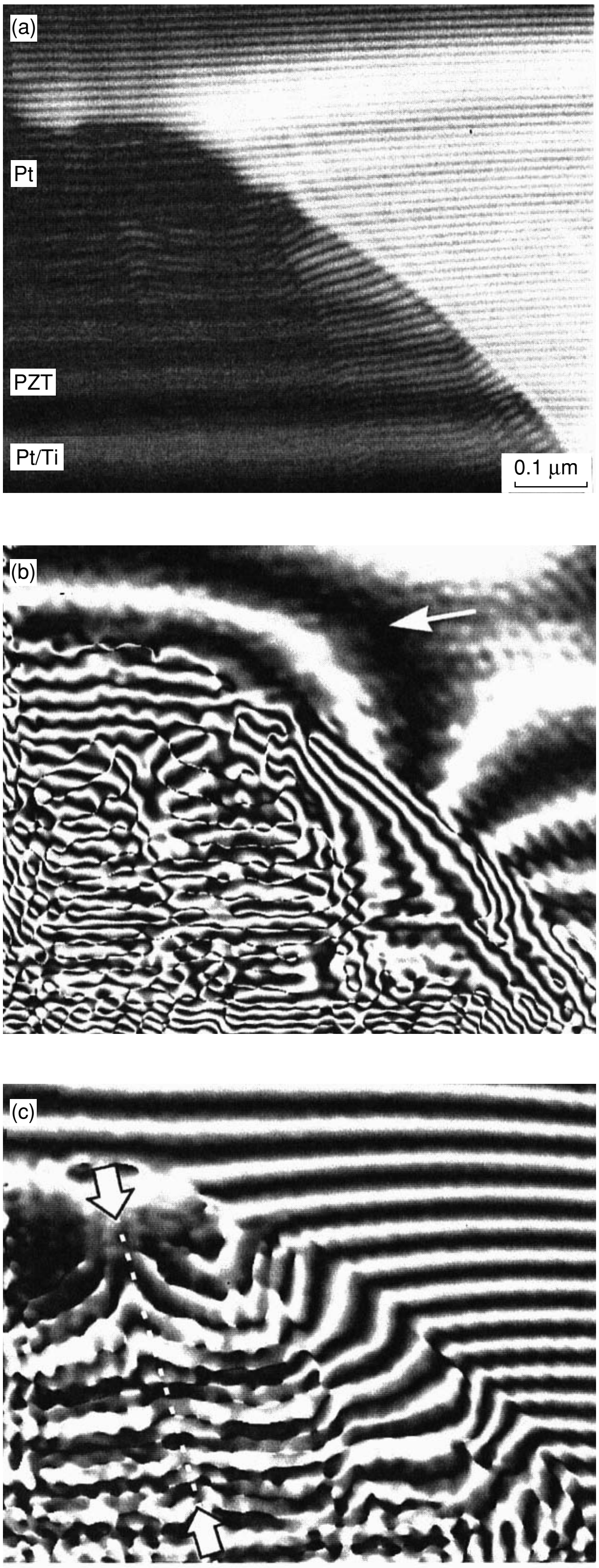

Figure 4
(a) Cross-sectional hologram of PZT capacitor of FeRAM. (b) Phase image of capacitor obtained from hologram shown in (a). (c) Interferogram of same area.

The poly-PZT grain structure is revealed by dark-bright contrasts in the inner layer of the PZT film. Also, the contour patterns (S2) in the inner layer of the poly-PZT grains reveal the grains' potential distributions. In one grain, the contours are bent at three points (e1, e2, and e3). Since the contours are lines of equi-potential, the normal direction of the contours indicates the polarization direction of the area. In this grain, the contours are bent at these points, indicating a variation of polarization direction. This reveals the existence of 90° domains in the grain. From the contour bending, the size of these 90° domains are between 50 and 60 nm.

3.3 Cross section of FeRAM capacitor

Figure 4 (a) shows a cross-sectional hologram of the PZT capacitor of an FeRAM, and **Figure 4 (b)** shows the corresponding phase image.

In Figure 4 (b), in the vacuum region adjacent to the capacitor indicated by the arrow, the electron phase changes show the electric field distribution due to the polarization of the capacitor PZT film. This indicates that the poly-PZT film of this capacitor behaves as an FeRAM.

An interferogram of the same area is shown in **Figure 4 (c)**. The dashed line shows the grain boundary of the poly-PZT film. The interferogram reveals a significant change in electric field across the grains of the poly-PZT film.

This is the first representation of the poly-PZT thin film of a ferroelectric memory device by means of electron holography.

4. Summary

This paper described how we used off-axis electron holography to observe the potential distribution across a ferroelectric PZT capacitor and observe the surrounding electric field created by the spontaneous polarization of the PZT thin film. We presented figures showing the equi-phase contours of the incident electron beam. Because the phase shift magnitude of the beam was shifted according to the electric field strength in the PZT thin film, these figures can be used to visualize the electric field. This is the first electron-holographic representation of the electric field of a poly-PZT thin film of a ferroelectric memory device.

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