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Correlation between grain size and domain size distributions in ferroelectric media for probe storage applications

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The relationship between grain size and domain size distributions has been studied by piezoelectric force microscopy in ferroelectric films with average grain size of 150 nm. As the ratio of domain size to grain size increases, the domain size deviation decreases in a $1/x^n$-type function, where n is 1.105. Extrapolation of the model shows that in order to obtain 10% domain size deviation in 1 Tbit/in.^{2} media, a grain size smaller than 14 nm is required. The obtained results imply that either nanograin or single crystalline/epitaxial films provide reliable domain distributions for probe storage applications. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2363942]

Ferroelectric domain imaging using contact-mode atomic force microscopy (AFM) has been widely used to study ferroelectric films or bulk crystals.[1–3] This nondestructive technique enables us to observe the domain configuration in the projection plane of the film at a nanometer scale. High-resolution AFM studies about the evolution of domains in thin films have been instructive with respect to microscopic information about fatigue[4,5] and switching dynamics.[6,7] Domain formation in ferroelectric thin films has been of particular interest in high density probe storage applications. In probe-based data storage[8] (PBDS) electrical pulses are applied to the probe to write individual domains. Uniform domain sizes and shapes are required for reliable writing/recording.

In preferentially (111) oriented[6] and epitaxial (001) lead zirconate titanate (PZT),[7] it was found that the domain size was linearly dependent on pulse voltage and logarithmically on pulse width. Material perfection influences the domain definition: In polycrystalline films, a domain may be made of a number of grains, each contributing to the average domain polarization. It has been found that the switched area in polycrystalline PZT was confined by grain boundaries.[9] Therefore, unwanted scattering of properties among domains is expected to appear when the number of grains per domain is decreased, i.e., when the domain size reaches the grain size. As such, it has been reported that grain misalignment in $SrBi_2TaO_9$ led to increased standard deviations of the average polarization signal when the capacitor size was decreased.[10]

In this letter, we highlight the role of grain boundaries on the domain formation by correlating domain size and grain size. The sample was a 50 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. The schematic diagram of the experimental setup was reported in a previous publication.[6]

Initially, the domains were oriented from top to bottom by applying +5 V to the AFM tip over a scan area of $5 \times 5\ \mu m^2$. This process is referred to as background poling. The writing of domains inside the background-poled area has been performed at pulse widths of 0.01–15 ms with a fixed pulse voltage of −15 V. An ac modulation voltage of 0.6 V_{pp} (peak to peak) at 17 kHz was used for domain imaging over a scan area of $1 \times 1\ \mu m^2$. The domain size was measured on the phase images by taking a geometric average of each domain. From this information, the standard deviation of the domain size was calculated for each writing condition. Dividing this value by the average domain size gave the relative standard deviation of the domain size σ_r. This relative deviation represents the degree of nonuniformity in domain size under the same writing conditions.

To explain further the inflection points and the trends of the domain size distribution with the pulse width, we performed a simple model calculation.[11] In the calculation, we assumed that the grain boundary acts as an electric shield, as well as a current path.[12,13] Therefore, domains cannot be written whenever the tip touches the grain boundary.[11]

Figure 1 shows the domain size and its relative standard deviation σ_r as a function of the pulse width. The standard deviation was calculated from a sampling of over 40 measurements per pulse width. In the experimental results of Fig. 1(a), the deviation decreases as the domain size increases. However, in the calculation results of Fig. 1(b), the deviation decreases at both extrema of pulse widths.

From the calculation results, the trend of the deviation implies that uniform domains are formed when the discrepancy between the domain size and the grain size is large, whereas nonuniform domains are formed when the domain size is similar to the grain size, i.e., when the probability of including a grain boundary is close to or equal to 1. These

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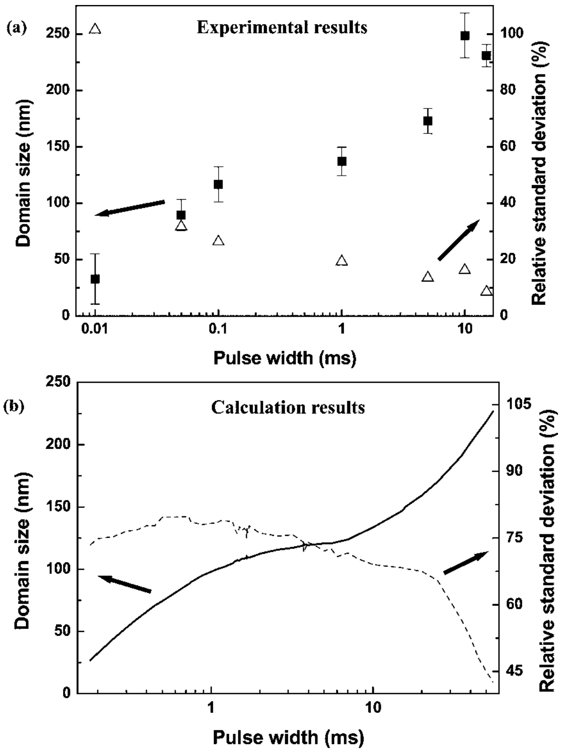

FIG. 1. Domain size and its relative standard deviation as a function of pulse width. Experimental results (a) and calculation (b).

three regions are schematically depicted in Fig. 2(a). If the domain size is much larger than the grain size [first region, Fig. 2(a)(1)], a large number of entire small grains form the domain. The border of the domain is constituted by the outer grain boundaries of the grains at the domain's perimeter. If the domain size is much smaller than the grain size (third region, Fig. 2(a)(3)], domains are mostly formed at the inner grain and a few domains are formed around grain boundaries. Finally, there is a region where the domain size is in the range of the grain size [second region, Fig. 2(a)(2)]. Here, in many cases, a section of the domain boundary is inside a grain where the shape is defined by the electric field and domain configuration, and another section is defined by the shape of the grain boundary. Therefore, in this region, highly nonuniform domains of different sizes are formed.

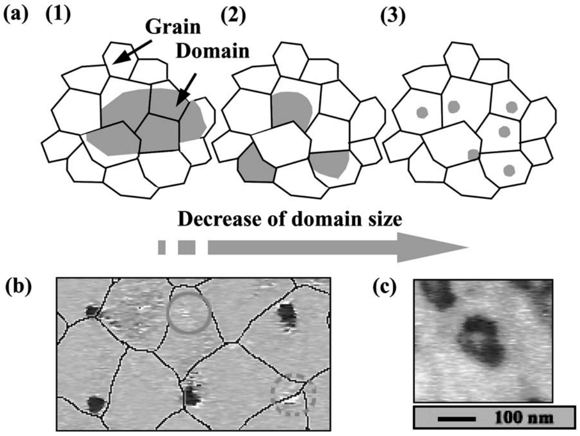

FIG. 2. (a) Schematic diagram of overlapped images of phase and topography image with different grain sizes and similar domain size. (b) Unwritten places in the overlapped images of phase and topography image. The line gray circle indicates the unwritten place inner grain and the dotted gray circle indicates the unwritten place around the grain boundary. (c) Polarization reversal inside the formed domain by a grounded tip.

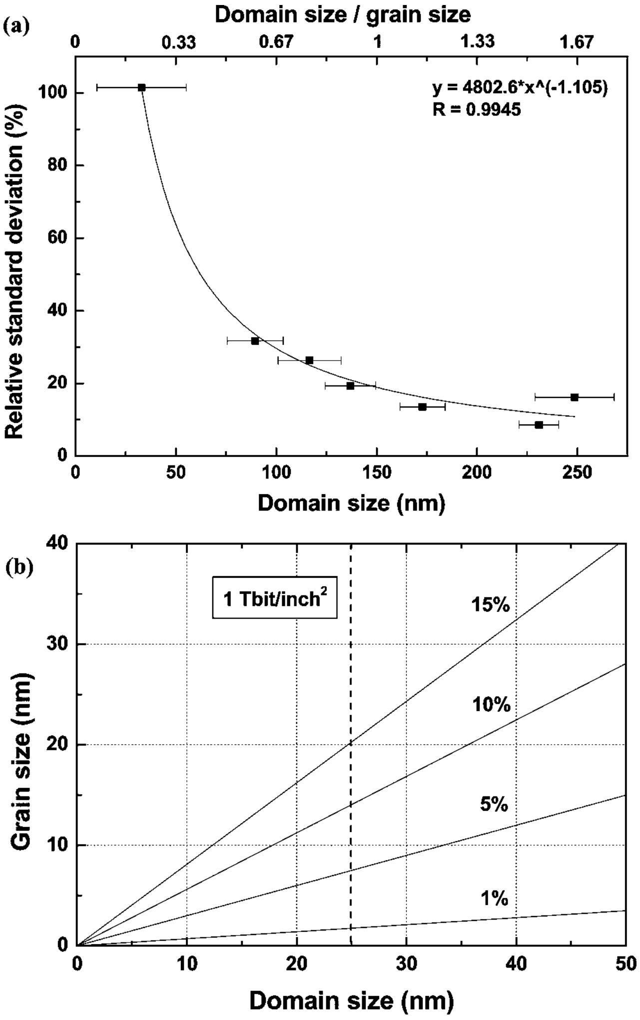

FIG. 3. (a) Domain size relative standard deviation as a function of domain size and a ratio of domain size over grain size for an average grain size of 150 nm. (b) Grain size vs domain size for constant σ_r.

Overall, the calculation results agree well with the experimental data. Only in the small pulse width region, the calculated relative deviation does not match well the experimental finding. Our model accounts for unwritten domains whenever the tip touches a grain boundary. However, unwritten domains have also been observed inside the grains [gray line in Fig. 2(b)]. There are several possible mechanisms for the unexpected phenomenon. One reason is different switching properties between each grain[14] due to imperfect (111) orientation.

Another reason is the polarization reversal by a grounded tip. As previously reported,[15] a grounded tip may induce polarization reversal if it is contacted on poled domains immediately after writing. In Fig. 2(c), the polarization reversal was observed by the appearance of a smaller, back-switched dot in the center of the written domain. The polarization reversal happens over a scale of several tens of nanometers. Therefore, in the region of small domain sizes, the polarization reversal leads to the complete back switching of the written dot.

Other reasons for unwritten domains may be defects inside the film, which disturb the movement of domain walls[16] and temporary loss of the tip-sample contact. For these reasons, the experimentally measured relative deviation is larger than the calculated one.

Obviously, the deviation is strongly affected by the ratio of domain size to grain size because of the number of grains incorporated into a domain. Figure 3(a) shows the relative standard deviation of the domain size σ_r as a function of the domain size and a ratio of domain size over grain size for an average grain size of 150 nm. From this figure, the decrease of σ_r with the domain size has been fitted to a $1/x^n$-type decay. This means that the model assumes infinite values for σ_r as the domain size approaches zero. The power index n has been found to be 1.105. Assuming that σ_r is constant for a given relative domain size (domain size/average grain size), we can estimate the necessary average grain size for a target value of σ_r. From this figure, at a given domain size, a small grain has a small deviation. Therefore, in order to have statistically uniform domain size distribution, one has to explore very small grain. Also, a single crystal film has statistically uniform domain size distribution due to the absence of the grain boundary. For example, for a PBDS system of 1 Tbit/in.[2] memory density, the domain size is about 25 nm. If σ_r is taken to 10%, the required grain size has to be below 14 nm. The relative standard deviation of domain size $\sigma_r = 1.0\%$ implies that only 4% of domains written at the same condition have a domain size which differs by more than 5% from the mean domain size. Graphs of constant σ_r are plotted in Fig. 3(b), showing the corresponding grain and domain sizes.

The above statements included only the grain size as a critical parameter for uniform domain sizes. However, a 25 nm large domain written in a film made of 14 nm sized grains contains an average of just four grains. Therefore, a large deviation in the domain shape can be expected. To circumvent that problem, the ferroelectric thin film should feature a narrow grain size distribution.

In conclusion, we found that the domain size distribution is dependent on the domain size. Uniform domain sizes can be written when the domain size is large compared to the grain size. The reason is that the domain size is less susceptible to the grain boundary. Therefore, the ultimate research direction for ferroelectric media development should be either nanograin (large number of grains per domain) or single crystal/epitaxial (no grain boundaries) thin films in order to ensure a reliable domain size distribution for PBDS.

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