The Origin of Local Strain in Highly Epitaxial Oxide Thin Films

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The ability to control the microstructures and physical properties of hetero-epitaxial functional oxide thin films and artificial structures is a long-sought goal in functional materials research. Normally, only the lattice misfit between the film and the substrate is considered to govern the physical properties of the epitaxial films. In fact, the mismatch of film unit cell arrangement and the Surface-Step-Terrace (SST) dimension of the substrate, named as “SST residual matching”, is another key factor that significantly influence the properties of the epitaxial film. The nature of strong local strain induced from both lattice mismatch and the SST residual matching on ferroelectric (Ba,Sr)TiO3 and ferromagnetic (La,Ca)MnO3 thin films are systematically investigated and it is demonstrated that this combined effect has a dramatic impact on the physical properties of highly epitaxial oxide thin films. A giant anomalous magnetoresistance effect ($\sim 10^{10}$) was achieved from the as-designed vicinal surfaces.

The lattice misfit between a hetero-epitaxial thin film and the substrate has a very strong influence on the crystal structure, microstructure, and physical properties of the deposited material. There has been a great deal of research on the influence of film strain in hetero-epitaxial films due to the film/substrate lattice misfit. The lattice misfit has been used to enhance the Curie temperature of high temperature superconductors, to alter the Curie temperature of ferromagnetic transition in Colossal Magnetoresistance (CMR) materials, and to change the dielectric properties of ferroelectric materials. In principle, the lattice misfit induced strain energy can be partially or fully released at the interface between the epitaxial film and substrates via formation of edge dislocations that periodically distribute along the interface. This has been directly observed and demonstrated by cross-sectional transmission electron microscopy (X-TEM) microstructural studies. However, only a few researches were reported for the investigation of the effect of the mismatch between the film unit cell arrangement and the substrate surface-step-terrace dimensions, named as “SST residual matching”, although some investigations have been done on the films grown on various vicinal substrate surfaces. It should be noted that there are a large number of surface-step-terraces on single crystal surfaces. Normally, a surface-step-terrace will not match an exact number of unit cells or atomic planes in the film, as seen in the Fig. 1. The mismatch of film unit cells/substrate terrace, SST residual matching, will result in an additional strain energy that cannot be released via edge dislocations and will be stored in the hetero-epitaxial films. As the film continues to grow away from the surface, the SST residual matching-induced strain energy will accumulate in the hetero-epitaxial film and can significantly alter its microstructure and thus, its physical and electrical properties. In fact, the effect of surface-step-terrace on the epitaxial nature of highly epitaxial oxide thin films can be considered in two factors: step height and terrace dimension. The step height can result in the formation of anti-phase domain boundaries (APB), conservative and non-conservative boundaries. However, the terrace dimension effect is normally neglected in thin film growth. Unlike metal or semiconductor thin films, ionic thin film growth requires a good match in the combination of positive and negative charge balance. Theoretically, when a number of film unit cells fill up along the terrace, a mismatch gap can be generated at the end of the step terrace, which is the “SST residual matching gap” or named as “mismatch gap”. Practically, this mismatch gap will not exist at the end of step terrace in an epitaxial film. The last atomic plane of the film will always occupy the atomic position of the terrace end.
rearrangement of the local atomic structure. Thus, the atomic position change can significantly increase the growth potential and alter the epitaxial quality.

We have investigated the effects of lattice mismatch on the dielectric properties of (Ba,Sr)TiO$_3$ (BSTO) and 2% Mn doped BSTO (Mn:BSTO) films$^{9,20}$. BSTO thin films have been extensively investigated as candidates for development of a new class of tunable microwave signal processing sensors and transducers as well as green energy harvest devices. The films exhibit an electric field dependent dielectric constant and have been used to fabricate tunable capacitors for use in microwave varactors and phase shifters. However, the relatively high loss tangent (tanδ) of BSTO films limits their use. It is observed that some materials exhibit a large electric field change in the dielectric constant, but also a high dielectric loss, while other materials exhibit a very small electric field effects, and at the same time a low dielectric loss. The challenge is to optimize the processing of the material such that it exhibits a large electric field change in tuning, while minimizing the dielectric loss$^{10,21,22}$.

CMR effect has been investigated for decade’s years$^{23–26}$, since it is a candidate for various electronic device applications, such as the read-head of magnetic memory, especially for the manganite materials and magnetic field sensor. On the other hand, it can provide valuable information to investigate magnetic field induced insulator-metal transition. Here, we report that high epitaxial BSTO and Mn:BSTO thin films on $1^\circ$, $3^\circ$ and $5^\circ$ vicinal (001) MgO substrate exhibit different dielectric constant and tunability induced by the SST residual mismatch. Also, the (La,Ca)MnO$_3$ thin film on vicinal (001) MgO substrate exhibits extremely high magnetoresistance ($\sim 10^9$), which is about four orders higher than the previous reported record. Thus, we successfully utilize film/substrate interactions to adjust the properties of the material for the desired application.

Results

To investigate the effect of the residual matching generated from the mismatch between the film unit cells and terrace dimension, we use various miscut (001) MgO substrates to create different vicinal surfaces for epitaxial growth of ferroelectric BSTO and Mn:BSTO thin films. The various miscut (001) MgO substrates can provide different surface-step-terrace dimensions on the vicinal surfaces. With miscut angles of $1^\circ$, $3^\circ$, and $5^\circ$ on the (001) vicinal MgO surfaces, the average step terrace dimension can be simply estimated to be 12.067 nm, 4.022 nm, and 2.413 nm, respectively. These dimensions correspond the average BSTO unit cells of 30.42, 10.17, and 6.06 on the $1^\circ$, $3^\circ$, and $5^\circ$ vicinal surface terraces. Fig. 2 gives dielectric property measurements from the epitaxial Mn:BSTO thin films grown on various miscut (001) MgO vicinal surfaces. Typical dielectric properties of both BSTO and Mn:BSTO thin films on these different miscut surfaces are summarized in Table 1 along with properties of films deposited on normal (001) MgO surface. Here, the tunability of the dielectric constant (i.e. change with an applied electric field) is defined in terms of the gap capacitance with a 0 V bias, and the gap capacitance with a 40 V bias, Tunability (%) = |(C(0 V) – C (40 V))/C (0 V)| × 100%.

As seen in Table 1, the dielectric constant and dielectric tunability of films grown on $1^\circ$ and $5^\circ$ miscut (001) MgO surfaces show very similar properties with the normal (0 degree miscut) substrates. However, both the dielectric constant and tunability for the film grown on $3^\circ$ miscut substrate are two-thirds of that for the films grown on $1^\circ$ and $5^\circ$ miscut (001) MgO surfaces. This dramatic difference in dielectric properties is due to the mismatch between the film unit cells in-plane and the terrace dimensions.

To better understand the effect of step terrace dimensions, X-TEM was employed to investigate the epitaxial quality and interface structures of the BSTO films on the (001) MgO vicinal substrates. Figs. 3 (a)–(c) are X-TEM images showing the epitaxial behavior and (d)–(f) are the interface structures for the BSTO films grown on $1^\circ$, $3^\circ$, and $5^\circ$ miscut substrate surfaces, respectively. The insets in Figs. 3 (a)–(c) are the selected area electron diffraction patterns (SAED) taken along the [100] zone of the MgO lattice. All three SAED patterns look similar, indicating that all of the films deposited on the miscut vicinal surfaces have good single crystallinity. The films grown on $1^\circ$ and $5^\circ$ miscut substrates reveal excellent hetero-epitaxy with very smooth surfaces and very sharp interfaces. Furthermore, edge dislocations are uniformly distributed along their entire interfaces. However, the film grown on the $3^\circ$ miscut (001) MgO substrate is very different. It is observed that the film consists of two layers: a highly epitaxial layer

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Figure 1 | Schematic illustration showing the surface-step-terrace dimension of the substrate can not exactly accommodate integer unit cells or atomic planes of the film.

Figure 2 | Dielectric property measurements showing the effects of surface step terrace induced local strain on the ferroelectric Mn:BSTO thin films grown on $1^\circ$, $3^\circ$, and $5^\circ$ miscut substrate surfaces, respectively.
on the miscut MgO substrate and a top polycrystalline-like layer. However, the electron diffraction pattern (inset in Fig. 3(b), clearly shows that all of the particle-like grains are well aligned along the c-axis. Although the interface between the epitaxial film and the MgO substrate is very sharp, the roughness of the film surface is high (as high as 50 nm). In addition, rather than the uniform dissemination, the edge dislocations along the interface are quasi-periodically distributed with a period close to 4.0 nm, as marked by arrows in Fig. 3(e). These phenomena can be understood by considering the unit cell arrangement on the substrate terraces.

As seen in Fig. 4(a), the film unit cells are orderly aligned on each terrace of the (001) MgO substrate surface. The surface terminations on the (001) MgO surface are always the MgO layer although each neighboring MgO layer has half unit cell height difference. Previous studies of BSTO films on (001) MgO have indicated that the TiO₂ layer is the nucleation layer of the film. Therefore, when BSTO grows on the (001) MgO substrate, the hetero-epitaxial BSTO film on each terrace becomes a single domain and the film consists of many domains which are shifted half unit cell along the c-axis if the neighboring terraces are single-step height terraces. The antidomain boundaries are therefore formed at the end of each step terrace. Details can be found in the literature. When the BSTO unit cells orderly align along a terrace, the number of BSTO unit cells is determined by the dimension of the terrace. It should be noted that the dimensions of a terrace are not equal to an exact numbers of BSTO unit cells. Theoretically, the difference of the match between the BSTO unit cells and terrace dimensions will generate a small space gap Δd on each terrace, as seen in Fig. 4(a). However, because of the lattice mismatch and the unit cell mismatch on the terrace, each terrace end in the epitaxial growth is always the atomic plane of the film, which results in the unit cells being reconstructed on the terrace, as shown in Fig. 4(b). This reconstruction results in the formation of local strain domain. The mismatch strain is completely dependent upon the mismatch of the film unit cells and terrace dimension d, or simply, the size of the mismatch gap Δd. The lattice mismatch induced strain can be defined as δ = Δd/d. Unlike the lattice mismatch in the film growth, the mismatch-induced strain cannot be released via formation of edge dislocations that generally occur in the lattice mismatch.

Thus, for the films grown on vicinal (001) MgO surfaces, the mismatch gaps can be estimated to be 0.42, 0.17, and 0.06 unit cells for the miscut of 1°, 3°, and 5°, respectively. For the film on 1°, it can’t be inserted the half unit cell of the BSTO film, since it will form no conservative antidomain. Thus, the strain of the BSTO film on the three vicinal substrates is tensile, but the average strain distribution in the 3° miscut is much larger than the film on the 1° and 5° miscut substrate. With film growth, the strain energy will be rapidly

| Sample Types | Residual Match Strain | εᵣ (0 V) | εᵣ (40 V) | εᵣ Tuning % | εᵣ (0 V) | εᵣ (40 V) | εᵣ Tuning % |
|--------------|----------------------|----------|----------|-------------|----------|----------|-------------|
| (001)-0.0° off | 0                    | 1480     | 755      | 49%         | 1068     | 712      | 33%         |
| (001)-1.0° off | 0.014                | 1664     | 800      | 52%         | 1028     | 682      | 34%         |
| (001)-3.0° off | 0.017                | 1059     | 663      | 37%         | 790      | 598      | 24%         |
| (001)-5.0° off | 0.010                | 1655     | 633      | 62%         | 1202     | 790      | 34%         |

Figure 3 | (a)–(c) are cross sectional-TEM images showing the epitaxial behavior and (d)–(f) are the interface structures for the BSTO films grown on 1°, 3°, and 5° miscut substrate surfaces, respectively. Edge dislocation distributions for the BSTO films grown on 1° and 5° (d and f) are periodic, but it is quasi-periodical on 3° (e). The inset of (a–c) is selection area of electron diffraction patterns (SAED) of the films taken along the [100] zone of the MgO lattice.
accumulated and stored in the films. When the strain accumulated in the film larger than the critical value for the film, the strain energy must be released with the form which will result in the alteration of the epitaxial behavior by forming the polycrystalline-like particles. This phenomenon is evident in the X-TEM image in the Fig. 3 (b) for the film on the 3° miscut substrate and can be well understood by growth dynamics under strain.

It is easy to understand the dielectric properties of the present films in terms of the observed structure. The dielectric measurements for the films on 1° and 5° miscut substrates display a higher dielectric constant and larger dielectric tunability than the film on the 3° miscut substrate in the in-planar interdigitated measurements. These results are somewhat consistent with the previous studies of the strain effects on BSTO thin films by capacitor measurements and (Ph,Sr)TiO3 (PSTO) thin films on (110) NdGaO3 (NGO) in in-planar interdigitated measurements. In the capacitor measurement, the BSTO films with compressive strain (in-plane) have much larger dielectric constant and higher dielectric tunability (out-plane) than those on the released ones (in-plane). We also found the dielectric constant and dielectric tunability in the tensile strain direction are 4220 and near 60% in ferroelectric PSTO thin films on (110) NGO substrates. These values are much greater than those of the films on the compressive strain directions with the dielectric constant value of only 1630 and 33%. Thus, the films on 1° and 5° miscut substrates with tensile strain have large dielectric constant and high dielectric tunability in the in-planar interdigitated measurements. Although the film on the 3° miscut substrate has the largest tensile strain, it has lowest dielectric constant and worst dielectric tunability possibly due to the fact when the strain is too large, it will distorted the crystal structure and generate more defect in the film. On the other hand, a polycrystalline-like particle structure and rough surface will also result in the increase of the dielectric loss and the reduction of the dielectric tunability. The boundary at polycrystalline-like particles and the interface between the highly epitaxial layer and the particle structural layer are usually oxygen deficient, even in the nano twinning lamellae structures.

To further verify the local strain formation and its effects on the physical properties, ferromagnetic La_{0.67}Ca_{0.33}MnO3 (LCMO) thin films were epitaxially grown on non-miscut and 5° miscut vicinal (001) MgO surfaces. Fig. 5 are the transport properties achieved from the non-miscut and 5° miscut (001) MgO surfaces. The film on the non-miscut MgO surface shows typical magnetoelectric properties with a clear ferromagnetic transition occurred at ~260 K. However, the film grown on the 5° miscut vicinal surface reveals an anomalous phenomenon where temperature dependence of resistivity is semiconductor behavior without the application of magnetic field, but under the magnetic field, temperature dependence of the resistivity measurements indicate that a ferromagnetic phase transition occurs at 115 K. Unlike the traditional phase transition for the ferromagnetic LCMO thin films on various substrates and its bulk material in which the transition temperature is increased with the applied magnetic fields, the transition does not change with the applied magnetic fields. Furthermore, a super-large magnetoresistance (MR) effect ratio of 10^4 was achieved from this system, which is about 4 orders higher than the early reported record of MR effect in this system. Details of the mechanisms are under investigation and will be reported later.

**Discussion**

This is a systematical study of the local strain effects induced by the film unit cell mismatch with surface step terraces on the physical properties of highly epitaxial oxide thin films. The lattice misfit and unit cell number/substrate terrace dimension mismatch were simultaneously considered. It is shown that this combined effect controls the epitaxial nature and has a dramatic impact on the physical properties of highly epitaxial oxide thin films. This remarkable finding can be utilized as a design tool in the growth of thin film materials for advanced applications.

**Methods**

A KrF excimer pulsed laser deposition with a wavelength of 248 nm was performed to deposit the three different thin films. The Ba0.6Sr0.4TiO3, 2% additional Mn doped Ba0.6Sr0.4TiO3, and La0.67Ca0.33MnO3 targets were used for the deposition. The BSTO and Mn:BSTO thin films were fabricated on 1°, 3°, and 5° miscut vicinal angles (001) MgO surfaces. The LCMO thin films were grown on non-miscut and 5° miscut vicinal (001) MgO surfaces.

The X-TEM was employed to understand the epitaxial quality and interface structures of the BSTO films on (001) MgO vicinal substrates. The dielectric properties (dielectric constant and loss tangent) were determined from interdigitated capacitors which were fabricated on top of the film by standard photolithography. A thin Cr adhesion layer (~100 Å) was used for the gold capped silver electrode. The silver was deposited in an e-beam evaporator over the patterned resist. Lift-off was then used to delineate the gap capacitor pattern. Thick silver (2–5 μm) was used to reduce the conductor losses at microwave frequencies (~1–20 GHz). The transport property measurements of the LCMO thin films were carried out with superconducting quantum interference device.

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Author contributions

C.M. had the magnetic measurements. M.L. had the sample preparation and assisted magnetic measurements. C.C. designed and setup the research and wrote the paper. Y.L. made the Mn:BSTO films and x-ray characterizations. Y.L. assisted manuscript preparation and data analysis. J.J. and E.I.M. had the TEM studies and microstructural analysis. Q.Y. assisted the data analysis. All authors discussed the results and commented on the manuscript.

Additional information

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