Anisotropy of crystal growth mechanisms, dielectricity, and magnetism of multiferroic Bi$_2$FeMnO$_6$ thin films

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Abstract
Epitaxial Bi2FeMnO6 (BFMO) thin films deposited on various Nb:SrTiO3 substrates show that the lattice parameters are very sensitive to epitaxial strains. Compressive and tensile strains are induced to the in-plane lattice constants of the (100) and (111) oriented films, respectively, while that of the (110) oriented thin film stay unstrained. The thin films also exhibit a strongly anisotropic growth habit depending on the substrate. Spiral growth, such as in the (100) BFMO film, is unique in samples prepared by pulsed laser deposition. Extrinsic dielectric constants at low frequencies are attributed to oxygen vacancies via the Maxwell-Wagner effect. All the samples show saturated hysteresis loops with very small coercive fields at 200 K, indicating the presence of weak ferromagnetism.

Keywords
crystal, growth, mechanisms, dielectricity, anisotropy, magnetism, films, multiferroic, bi2femno6, thin

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Epitaxial Bi$_2$FeMnO$_6$ (BFMO) thin films deposited on various Nb:SrTiO$_3$ substrates show that the lattice parameters are very sensitive to epitaxial strains. Compressive and tensile strains are induced to the in-plane lattice constants of the (100) and (111) oriented films, respectively, while that of the (110) oriented thin film stay unstrained. The thin films also exhibit a strongly anisotropic growth habit depending on the substrate. Spiral growth, such as in the (100) BFMO film, is unique in samples prepared by pulsed laser deposition. Extrinsic dielectric constants at low frequencies are attributed to oxygen vacancies via the Maxwell-Wagner effect. All the samples show saturated hysteresis loops with very small coercive fields at 200 K, indicating the presence of weak ferromagnetism. © 2013 American Institute of Physics
cubic BiFeO₃, the out-of-plane lattice parameter of epitaxial BFMO thin films. The inset of (a) shows a quality image of a 1 nm thick film, presenting the slowest growth at the spiral centre, and faster growth as the distance from the spiral centre increases. It is evident that terrace layer growth is absent from the BFMO (100) thin film. The differences can be understood by comparing the dissimilarities of the crystalline conditions. In conventional spiral growth, aqueous solutions allow atoms to be uniformly absorbed around SDs after effectively moving, and they then go on to create atomically flat terraces. For PLD, atoms contained in the plasma are locally confined to the area where they reach the substrate surface. When a SD intersects the surface, a depression forms the slowest growth at the spiral centre, and faster growth as the distance from the spiral centre increases. It is evident that terrace layer growth is absent from the BFMO (100) thin film. The differences can be understood by comparing the dissimilarities of the crystalline conditions. In conventional spiral growth, aqueous solutions allow atoms to be uniformly absorbed around SDs after effectively moving, and they then go on to create atomically flat terraces. For PLD, atoms contained in the plasma are locally confined to the area where they reach the substrate surface. Atoms tend to simultaneously crystallize around the closest defects to form spiral ridges.

The FESEM surface morphologies shown in Fig. 3 are consistent with the observations from AFM images. Spiral growth was doubly confirmed in the BFMO (100) thin film. Stripe structures were also supported by the SEM surface morphology of the BFMO (110) sample. More exact surface details for the BFMO (111) film were obtained from the FESEM images. Small grains actually exist in the shape of pyramids, suggesting the [111] direction of epitaxial growth. Large grains are linked to each other at an angle of 120°, forming hexagons. It is important to check for composition differences between large and small grains in such a case. The EDS results show that their compositions are identical, supporting the absence of any impurity peaks in the XRD patterns.
patterns. Thus, large hexagons are caused by disequilibrium crystallization due to lattice mismatch between the thick overlay and the substrate.

The frequency-dependent relative dielectric constant and dielectric loss of the thin films are shown in Fig. 4. The relative dielectric constants decrease monotonically with increasing frequency, but only slightly at frequencies higher than 30 kHz. The relative dielectric constant values at 1 MHz are 130, 56, and 36 for BFMO (100), BFMO (111), and BFMO (110), respectively. Dielectric losses also show similar decreasing trends with increasing frequency, except that they display peaks below 200 kHz. The corresponding dielectric losses at 1 MHz are 0.34, 0.38, and 0.20. Our dielectric data on the BFMO thin films are in good agreement with the previous reports on BFMO ceramics. As shown in Fig. 4, an dielectric relaxation process is observed in the dielectric loss measurements below 200 kHz, but it is absent from the dielectric constant data over that range of frequency. This observation further demonstrates that extrinsic dielectric values at low frequencies could be attributed to the Maxwell-Wagner effect, namely, interfacial polarization generated by quasi-mobility of locally uncompensated space charges.

Fig. 5 presents the in-plane isothermal magnetization curves of the samples at 200 K. All the samples show saturated hysteresis loops with very small coercive fields, indicating the presence of weak ferromagnetism. The magnetizations at 5000 Oe are 0.057 emu/mm³, 0.078 emu/mm³, and 0.109 emu/mm³, corresponding to the BFMO (100), BFMO (110), and BFMO (111) thin films, respectively. Thus, magnetic anisotropy is induced by substrate strains via the magneto-elastic effect. Our observations are similar to those in other reports, but with relatively larger magnetizations.

In conclusion, epitaxial BFMO thin films of single phase were successfully prepared by the PLD method on (100), (110), and (111) NSTO substrates, respectively. Strain effects give rise to completely different surface morphology features. In particular, the spiral growth observed in the BFMO (100) thin film is due to formation of SDs in the process of deposition. All the samples show weak ferromagnetism at 200 K indicated by saturated hysteresis loops with very small coercive fields. The extrinsic dielectric values at low frequencies could be attributed to the Maxwell-Wagner effect.

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