Epitaxial growth of single BiFeO₃ films by rf magnetron sputtering

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Multifunctional oxide materials such as $BiFeO_3$ (BFO), $BiMnO_3$ and $YMnO_3$, etc have been widely investigated in recent years because they exhibit a coexistence of (anti-)ferromagnetic and (anti-)ferroelectric phases. Thus, they provide an ability to control the electric polarization by means of a magnetic field and the magnetization by an electric field. These properties make them attractive candidates for novel device applications in the emerging field of spintronics.[1-3] However, it is very important that these properties should be exhibited at room temperature for application in devices operating at the same. Although, to date a large number of magnetoelectric materials such as BFO [Neltemperature $(T_N \sim 643$ K)],[4] $YMnO_3$ $(T_N = 70 \sim 100$ K),[5] $HoMnO_3$ $(T_N \sim 75$ K),[6] and $TbMn_2O_5$,[7] etc have been reported, only BFO has been shown to possess not also a high magnetic curie temperature $(T_N \sim 643$ K) but also weak ferromagnetism at room temperature and a very high ferroelectric Curie temperature $(T_C \sim 1103$ K). In addition, it has been reported recently by J. Wang et al. that the application of an in-plane compressive stress in BFO films causes an order of magnitude enhancement in the spontaneous polarization as well as a large saturation magnetization (M_S) compared to bulk BFO at room temperature.[4] These studies provide the possibility of fabricating new types of devices in which electric polarization can be controlled by means of an applied magnetic field and vice versa. However, most epitaxial BFO films reported in literature to date have been obtained only by Pulsed Laser Deposition (PLD).[4,8-11] Although PLD is one of the most effective methods for the deposition of such multicomponent oxide materials,it is still not an industrially viable process because of the difficulties associated with producing large area films. In this context we have successfully deposited epitaxial single BFO films on (001) $SrTiO_3$ (STO) using the O_2 reactive rf magnetron sputtering technique. This paper describes the deposition conditions, crystal structure, microstructure and the magnetic properties of epitaxial BFO films obtained using the sputtering technique.

The (001) oriented BFO films with a thickness of about 200 nm were sputtered from a commercially available stoichiometric single BFO target on to single crystal (001) STO substrates. STO is cubic with the lattice parameter, a = 3.905 Å and has a misfit of 1.4 % with BFO. The crystalline quality of our films was investigated using X-ray diffraction (XRD) as well as rocking-curve analysis. Impurities such as iron oxides and bismuth oxides are seen in the XRD data, when conditions are not optimized.

We have also measured the magnetic hysteresis curves at room temperature as well as at 5 K using a superconducting quantum interface device (SQUID) magnetometer. The saturation

magnetization (M_S) values in both cases is nearly the same and is about 0.8 B/F.U. This value is almost the same as that reported by J. Wang et al.[4] However, it is an order of magnitude smaller than that reported by W. Eerenstein et al.[12]A key to understand the origin of such a difference is the presence of oxygen vacancies. It is well known that presence of oxygen vacancies in BFO results in the mixed valency of Fe and hence, an equivalent amount of Fe^{3+} goes in to the Fe^{2+} state to preserve charge neutrality. The presence of Fe^{2+} ions may give rise to a ferromagnetic exchange interaction of the type $Fe^{2+} - O^{2-} - Fe^{3+}$ and this may be the reason for the large values of M_S.

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