

Magnetic Structure of BiFeO₃ thin films

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Multiferroic materials have enjoyed a renaissance with the discovery of new materials such as TbMnO₃, TbMn₂O₅, and Ca₃CoMnO₆ [1]. However, despite the rich physics found in such compounds, their ordering temperatures are too low for device applications. Fortunately, BiFeO₃ (BFO) has both ferroelectric and magnetic order at room temperature, and it has been shown that electric fields can affect the magnetic domain populations of both single crystal and thin film materials. While the magnetic structure has been determined in single crystals to be a long wavelength spiral [2], in thin films it has been found to be different. In rather thin films (tens to a few hundred nm), the spiral collapses to form a G-type antiferromagnet [3]. In thicker films (of 1 micron), it forms a modulated structure [4].

Our BFO thin films are grown on (001) SrTiO₃ substrates. The thin film is 1 μm-thick and belongs to the “thicker” class of films in which the magnetic structure is a spiral or amplitude modulated. The BFO in these films have a monoclinic structure with $a = 5.645 \text{ \AA}$, $b = 5.589 \text{ \AA}$, $c = 3.950 \text{ \AA}$ and $\beta = 90.85^\circ$. One sample has been poled (-20 V) with an electric field and consists of a single ferroelectric domain. Our earlier experiments collected at room temperature on the BT9 instrument at NIST established [4] that the BFO thin film consists of a single magnetic domain with the magnetic moments lying in the (hhl) plane with a propagation vector along the [1,1,-2] direction. We found a modulated structure. Constraints on the plane of the moments were determined through polarized beam experiments performed on the BT7 spectrometer using ³He polarizers. In these polarized experiments, increasing inhomogeneity in the spin transport prevented us from uniquely determining the moment direction and we were only able to investigate the nature of magnetic scattering at satellite peaks about the (0.5 0.5 0.5) reflection. Furthermore, both satellites survived in both spin flip channels. While this would suggest an amplitude modulated structure, it cannot rule out the presence of equally populated chirality domains.

Using the N5 triple axis spectrometer, we first measured the reciprocal space map in the zone defined by the (111) and (1-10) reflections. The results [Fig. 1(a)] showed two peaks, indicating an incommensurate structure with a periodicity comparable to that of single-crystal samples. From careful fits to a ψ scan through the center of the two reflections [Fig. 1(b)], we found a ~3:1 in-plane/out-of-plane contribution ratio to the observed peaks.

To further investigate the magnetic structure of BFO thin films, we employed the polarized setup at the C5 triple axis spectrometer. The neutron beams were polarized with Heusler (111) crystals as monochromator and analyzer. Neutron polarization was maintained in the incident and scattered beam channel using permanent magnet guide fields. To ensure higher order contaminations are eliminated from the beam, two PG filters were placed after the sample. Mezei flippers were placed before and after the sample to allow the full measurement of all four scattering cross-sections (I^{++} , I^{-} , I^{+-} , I^{+}). A 5-coil Helmholtz assembly was used to control the neutron spin-orientation at the sample position by producing a magnetic field of order of 10 G. The orientation of the magnetic field at the sample position was automatically adjusted to allow the measurements to be performed for the neutron spin perpendicular to the scattering plane as well as parallel to any orientation in the scattering plane including parallel to the momentum transfer. The flipping ratio was measured to be 15:1 for various field configurations.

Figure 1 shows the results [5] of our polarized beam measurements at the (0.5 0.5 0.5) position in the zone defined by the (111) and (1-10) reflections. For $P \parallel Q(0.5 0.5 0.5)$, splitting between the +/− and −/+ cross sections was strong, consistent with a chirality axis along the [111] direction and magnetic moments in the plane normal to [111]. For $P \perp Q$, the two spin-flip cross sections gave the same intensity [Fig. 3(d)], also consistent with a chirality axis along [111]. Thus, we conclude that the magnetic structure of this BFO film

differs markedly from that of the single crystal. There is a cycloid with moments spiraling in the plane normal to the polarization, consistent with theoretical predictions of an easy plane in films [6].

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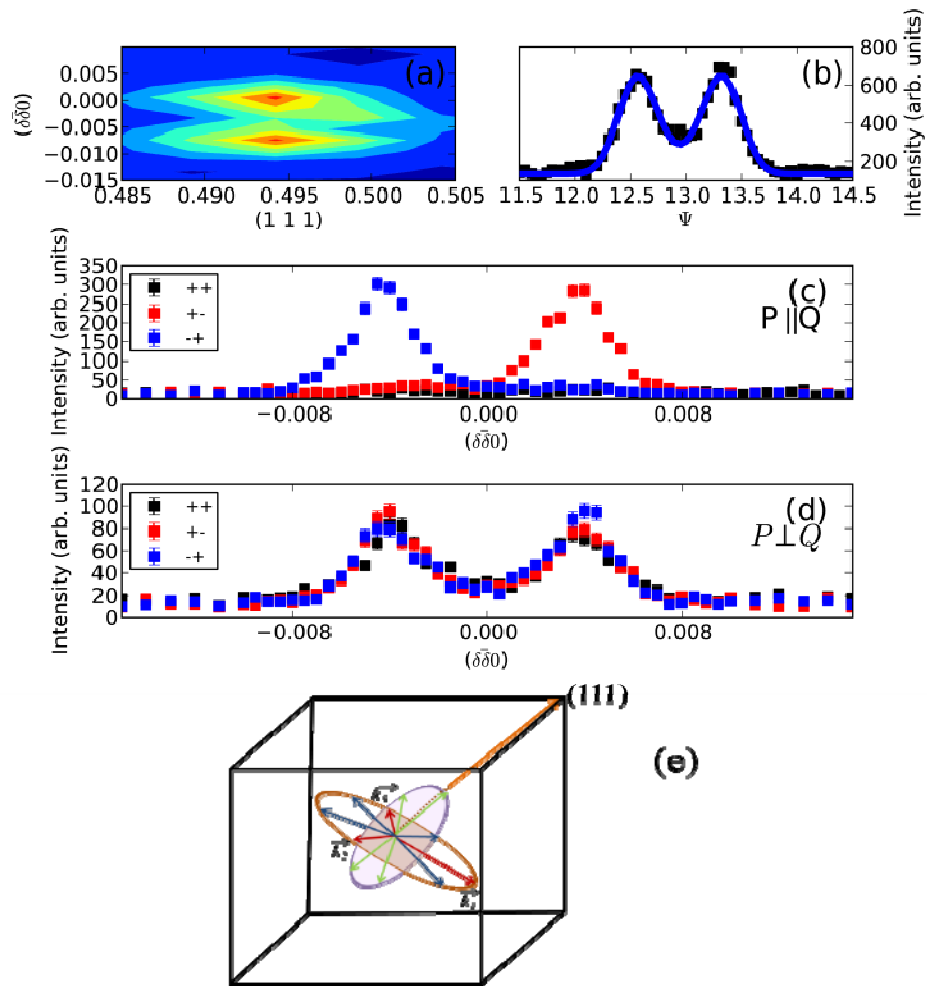


Figure 1 Contour maps of the neutron diffraction in the $(H\ K\ H+K/2)$ zone for the sample about the $(0.5\ 0.5\ 0.5)$ position in reciprocal space for: (a) after the application of -200kV/cm . (b) A ψ cut through the two peaks (c) SF and NSF cross-sections for $P \parallel Q$ measured about the $(0.5\ 0.5\ 0.5)$ position. (d) SF and NSF cross-sections for $P \perp Q$ measured about the $(0.5\ 0.5\ 0.5)$ position. (e) Cartoon of the magnetic structure. The red arrows denote the propagation wave-vectors for the 3 magnetic domains possible in the system. The red plane shows the plane of spiral for this film. Blue arrows denote moments in this plane. The purple plane represents the plane of the spiral determined for single crystals (for a domain defined by the \vec{k}_3 propagation vector.). Green arrows denote moments in this plane. Figure 1 is taken from our published paper [5].