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Randy S. Fishman

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# Pinning, Rotation, and Metastability of BiFeO<sub>3</sub> Cycloidal Domains in a Magnetic Field \*

Randy S. Fishman

Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, USA

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Earlier models for the room-temperature multiferroic BiFeO<sub>3</sub> implicitly assumed that a very strong anisotropy restricts the domain wavevectors  $\mathbf{q}$  to a three-fold symmetric axis normal to the static polarization  $\mathbf{P}$ . However, recent measurements demonstrate that the domain wavevectors  $\mathbf{q}$  rotate within the hexagonal plane normal to  $\mathbf{P}$  away from the field orientation  $\mathbf{m}$ . We show that the previously neglected three-fold anisotropy  $K_3$  restricts the wavevectors to lie along the three-fold axis in zero field. Taking  $\mathbf{m}$  to lie along a three-fold axis, the domain with  $\mathbf{q}$  parallel to  $\mathbf{m}$  remains metastable below  $B_{c1} \approx 7$  T. Due to the pinning of domains by non-magnetic impurities, the wavevectors of the other two domains start to rotate away from  $\mathbf{m}$  above 5.6 T, when the component of the torque  $\boldsymbol{\tau} = \mathbf{M} \times \mathbf{B}$  along  $\mathbf{P}$  exceeds a threshold value  $\tau_{\text{pin}}$ . Since  $\boldsymbol{\tau} = 0$  when  $\mathbf{m} \perp \mathbf{q}$ , the wavevectors of those domains never become completely perpendicular to the magnetic field. Our results explain recent measurements of the critical field as a function of field orientation, small-angle neutron scattering measurements of the wavevectors, as well as spectroscopic measurements with  $\mathbf{m}$  along a three-fold axis. The model developed in this paper explains how the three multiferroic domains of BiFeO<sub>3</sub> for a fixed  $\mathbf{P}$  can be manipulated by a magnetic field.

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## I. INTRODUCTION

The manipulation of magnetic domains with electric and magnetic fields is one of the central themes [1–3] in the study of multiferroic materials. Applications of multiferroic materials depend on a detailed understanding of how domains respond to external probes. Despite recent advances [4] in our understanding of the room-temperature multiferroic BiFeO<sub>3</sub>, however, some crucial questions remain about how its cycloidal domains respond to a magnetic field.

A type I or “proper” multiferroic, BiFeO<sub>3</sub> exhibits a strong ferroelectric polarization of about 80  $\mu\text{C}/\text{cm}^2$  along one of the pseudo-cubic diagonals below the ferroelectric transition at  $T_{FE} = 1100$  K [5,6]. Below  $T_{FE}$ , broken symmetry produces two Dzyaloshinskii-Moriya (DM) interactions between the  $S = 5/2$  Fe<sup>3+</sup> ions. A magnetic transition at  $T_N = 640$  K [7] allows the cycloidal spin state to take advantage of this broken symmetry.

Until recently, it seemed that a complete theoretical description [8–13] of rhombohedral BiFeO<sub>3</sub> was in hand. Utilizing the first available single crystals, the measured

cycloidal frequencies [14–16] of BiFeO<sub>3</sub> provided a stringent test for theory. A microscopic model for BiFeO<sub>3</sub> that includes two DM interactions  $D_1$  and  $D_2$  and single-ion anisotropy  $K_1$  successfully predicted [12,13] the mode frequencies in zero field [14,15] and their evolution in magnetic field [16] for several field orientations. Since all model parameters were determined [17] from the zero-field behavior of BiFeO<sub>3</sub>, the field evolution of the cycloidal modes [13] provided a particularly good test of the microscopic model. But new evidence suggests that this model is not complete.

With the electric polarization  $\mathbf{P} = P\mathbf{z}'$  along the pseudo-cubic diagonal  $[1, 1, 1]$  ( $[a, b, c]$  is a unit vector), the three magnetic domains of BiFeO<sub>3</sub> in zero field [18] have wavevectors  $\mathbf{Q}_k = \mathbf{Q}_0 + \mathbf{q}_k$  where  $\mathbf{Q}_0 = (\pi/a)(1, 1, 1)$  is the antiferromagnetic (AF) Bragg vector,

$$\mathbf{q}_1 = \frac{2\pi\delta}{a}(-1, 1, 0), \quad (1)$$

$$\mathbf{q}_2 = \frac{2\pi\delta}{a}(1, 0, -1), \quad (2)$$

$$\mathbf{q}_3 = \frac{2\pi\delta}{a}(0, -1, 1), \quad (3)$$

$a = 3.96$  Å is the lattice constant of the pseudo-cubic unit cell, and  $\delta \approx 0.0045$  determines the cycloidal wavelength  $\lambda = a/\sqrt{2}\delta \approx 620$  Å. As shown in Fig.1, each  $\mathbf{q}_k$  lies along a different hexagonal axis perpendicular to  $\mathbf{z}'$ . In zero field, the three domains of BiFeO<sub>3</sub> with wavevectors  $\mathbf{q}_k$  are degenerate. For each domain  $k$ , the spins of the cycloid lie primarily in the plane defined by  $\mathbf{z}' = [1, 1, 1]$  and  $\mathbf{x}'$ , which is the unit vector along  $\mathbf{q}_k$ . A magnetic field favors domains with  $\mathbf{x}' \perp \mathbf{B}$  because  $\chi_{\perp} \gg \chi_{\parallel}$  [19] for BiFeO<sub>3</sub>.

The model described above successfully predicted the

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TABLE I: Reference frames of BiFeO<sub>3</sub>

unit vectors	description and values
$\{\mathbf{x}, \mathbf{y}, \mathbf{z}\}$	pseudo-cubic unit vectors $\mathbf{x} = [1, 0, 0]$ , $\mathbf{y} = [0, 1, 0]$ , $\mathbf{z} = [0, 0, 1]$
$\{\mathbf{x}', \mathbf{y}', \mathbf{z}'\}$	rotating reference frame of cycloid $\mathbf{x}' \parallel \mathbf{q}$ , $\mathbf{z}' = [1, 1, 1]$ , $\mathbf{y}' = \mathbf{z}' \times \mathbf{x}'$
$\{\mathbf{X}, \mathbf{Y}, \mathbf{Z}\}$	fixed reference frame of hexagonal plane $\mathbf{X} = [1, 0, -1]$ , $\mathbf{Y} = [-1, 2, -1]$ , $\mathbf{Z} = [1, 1, 1]$

These DM interactions are created by the antiferrodistortive and polar modes of the BiFeO<sub>3</sub> lattice [29–32] that develop below its ferroelectric transition temperature.

The first DM term in  $\mathcal{H}$ ,

$$\mathcal{H}_{D_1} = D_1 \sum_{\langle i,j \rangle} (\mathbf{z}' \times \mathbf{e}_{i,j}/a) \cdot (\mathbf{S}_i \times \mathbf{S}_j), \quad (5)$$

does not depend on the choice of domain and  $\mathbf{q}_k$ . In earlier [4] versions of the “canonical” model, this term was restricted to a specific domain of the cycloid. For domain 2 with  $\mathbf{q}_2$  parallel to  $\mathbf{x}' = [1, 0, -1]$  and  $\mathbf{y}' = \mathbf{z}' \times \mathbf{x}' = [-1, 2, -1]$ , it was written

$$\mathcal{H}'_{D_1} = -\frac{D_1}{\sqrt{2}} \sum_{\mathbf{R}_j = \mathbf{R}_i + a(\mathbf{x} - \mathbf{z})} \mathbf{y}' \cdot (\mathbf{S}_i \times \mathbf{S}_j), \quad (6)$$

where  $\mathbf{R}_i$  and  $\mathbf{R}_j$  are next-nearest neighbors of the pseudo-cubic unit cell that lie on the same hexagonal layer  $h_i$ . This earlier version of the “canonical” Hamiltonian  $\mathcal{H}'$  implicitly assumed that the anisotropy is so high that the domain wavevector  $\mathbf{q}$  is restricted to one of the three-fold axis. Because the wavelength of the cycloid is so long,  $\mathcal{H}'$  has the same static and dynamical properties as  $\mathcal{H}$  provided that  $\mathcal{H}$  is applied to the domain specified by  $\mathcal{H}'_{D_1}$ .

Why replace  $\mathcal{H}'$  with  $\mathcal{H}$ ? Unlike  $\mathcal{H}'$ ,  $\mathcal{H}$  can be used to study any domain with  $\mathbf{q}_k$  along a three-fold axis. As shown below,  $\mathcal{H}$  also describes the general case where  $\mathbf{q}$  differs from a three-fold axis. While  $\mathcal{H}'_{D_1}$  involves the sum over next-nearest neighbors,  $\mathcal{H}_{D_1}$  involves the sum over nearest neighbors, which should dominate the DM interaction. Finally, the general form of  $\mathcal{H}_{D_1}$  given above was obtained from first-principles calculations [33].

To construct the local reference frame of a cycloid with wavevector  $\mathbf{q} = \mathbf{Q} - \mathbf{Q}_0$ , we take

$$\mathbf{q} = \frac{2\sqrt{2}\pi\delta}{a|\mathbf{n}|} (n_x, n_y, n_z) = \frac{2\pi}{\lambda} \mathbf{x}', \quad (7)$$

where  $n_i$  are integers with no common factors. Then the unit vector along  $\mathbf{q}$  is  $\mathbf{x}' = (n_x, n_y, n_z)/|\mathbf{n}|$  and  $\mathbf{y}' = \mathbf{z}' \times \mathbf{x}' = (n_z - n_y, n_x - n_z, n_y - n_x)/(\sqrt{3}|\mathbf{n}|)$ . With the local reference frame of a cycloid defined by the unit vectors  $\{\mathbf{x}', \mathbf{y}', \mathbf{z}'\}$ , the spin at site  $\mathbf{R}_i = (l, m, o)a$  is indexed by the integer  $r = \mathbf{n} \cdot \mathbf{R}_i/a = n_x l + n_y m + n_z o$ . Assuming that the spins  $\mathbf{S}_r^{(j)}$  on alternate layers  $j = 1$  or  $2$  are

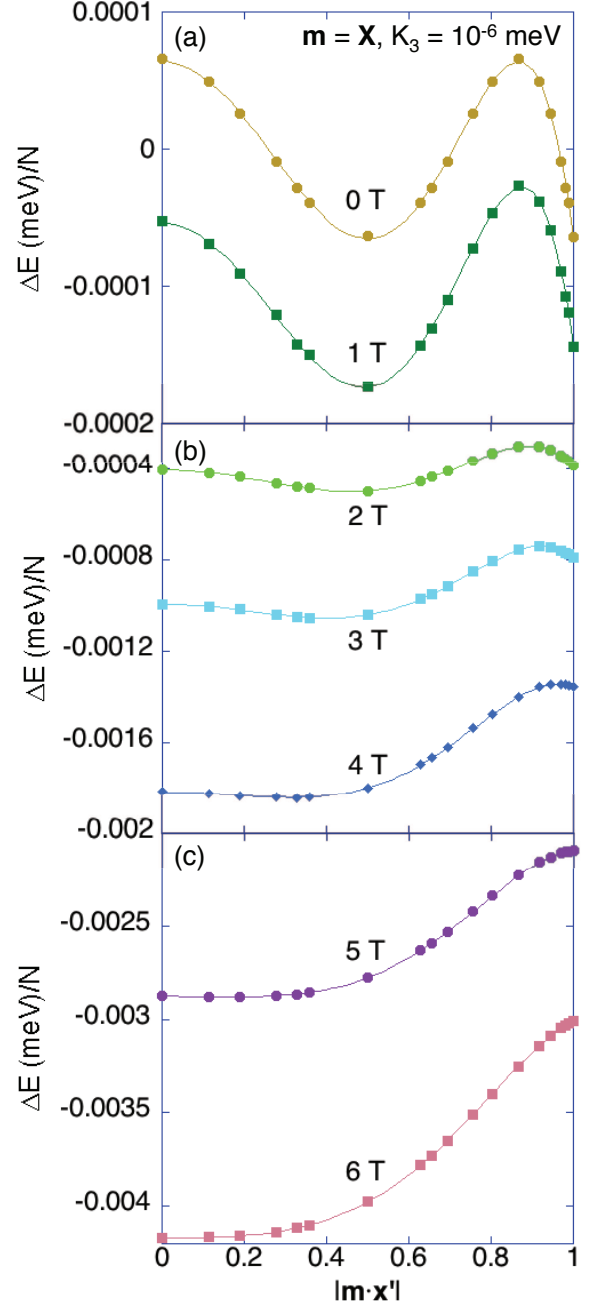


FIG. 2: (Color online) The energy  $\Delta E/N$  versus wavevector  $|\mathbf{m} \cdot \mathbf{x}'|$  for  $K_3 = 10^{-6}$  meV and field ranging from 0 to 6 T along  $\mathbf{X}$ . Solid curves are fits to a sixth order polynomial in  $|\mathbf{m} \cdot \mathbf{x}'|$

identical functions of  $r$ , then  $r$  ranges from 1 to  $M = |\mathbf{n}|/\sqrt{2}\delta = \lambda|\mathbf{n}|/a$  in the magnetic unit cell.

It is straightforward to show that

$$\frac{1}{N}\mathcal{H}_{D_1} = \frac{D_1}{2\sqrt{3}M} \sum_{r=1}^M \left\{ \begin{aligned} & \mathbf{x} \cdot \left[ \mathbf{S}_r^{(1)} \times (\mathbf{S}_{r+n_z}^{(2)} - \mathbf{S}_{r-n_z}^{(2)} - \mathbf{S}_{r+n_y}^{(2)} + \mathbf{S}_{r-n_y}^{(2)}) \right] \\ & + \mathbf{y} \cdot \left[ \mathbf{S}_r^{(1)} \times (\mathbf{S}_{r+n_x}^{(2)} - \mathbf{S}_{r-n_x}^{(2)} - \mathbf{S}_{r+n_z}^{(2)} + \mathbf{S}_{r-n_z}^{(2)}) \right] \\ & + \mathbf{z} \cdot \left[ \mathbf{S}_r^{(1)} \times (\mathbf{S}_{r+n_y}^{(2)} - \mathbf{S}_{r-n_y}^{(2)} - \mathbf{S}_{r+n_x}^{(2)} + \mathbf{S}_{r-n_x}^{(2)}) \right] \end{aligned} \right\}. \quad (8)$$

Since  $\mathbf{S}_{r+M}^{(j)} = \mathbf{S}_r^{(j)}$ , the index  $r + n_\alpha$  can be taken mod  $M$  to lie between 1 and  $M$ . Because  $\lambda/a = M/|\mathbf{n}| \gg 1$ ,  $|n_i| \ll M$  and

$$\mathbf{S}_{r+n_i}^{(2)} - \mathbf{S}_{r-n_i}^{(2)} \approx n_i (\mathbf{S}_{r+1}^{(2)} - \mathbf{S}_{r-1}^{(2)}), \quad (9)$$

with corrections of order  $\delta^3 \sim 10^{-7}$ . This leads to the simpler form [34]

$$\frac{1}{N}\mathcal{H}_{D_1} = \frac{D_1|\mathbf{n}|}{2M} \mathbf{y}' \cdot \sum_{r=1}^M \left\{ \mathbf{S}_r^{(1)} \times (\mathbf{S}_{r+1}^{(2)} - \mathbf{S}_{r-1}^{(2)}) \right\}. \quad (10)$$

Hence, the first DM interaction produces a cycloid in the  $\mathbf{x}' - \mathbf{z}'$  plane for any wavevector  $\mathbf{q} \parallel \mathbf{x}'$ .

The second DM interaction, can be similarly written as [28,33]

$$\frac{1}{N}\mathcal{H}_{D_2} = \frac{3D_2}{M} \mathbf{z}' \cdot \sum_{r=1}^M (\mathbf{S}_r^{(1)} \times \mathbf{S}_r^{(2)}), \quad (11)$$

which rotates alternate layers of spins about the  $\mathbf{z}'$  axis and tilts the cycloid out of the  $\mathbf{x}' - \mathbf{z}'$  plane

Neither of these DM interactions fixes the orientation of  $\mathbf{q}$  along a three-fold axis in zero field! By replacing  $\mathcal{H}'$  with  $\mathcal{H}$  in order to set the domain wavevectors  $\mathbf{q}$  free from the three-fold axis, we have eliminated all sources of anisotropy within the hexagonal plane. To remedy that deficiency, we must add an additional term to the Hamiltonian that breaks the three-fold symmetry in the hexagonal plane perpendicular to  $\mathbf{z}'$ .

### III. ANISOTROPY ENERGIES

Because  $\{\mathbf{x}', \mathbf{y}', \mathbf{z}'\}$  already provides the reference frame for the cycloid, which can rotate in the plane perpendicular to  $\mathbf{z}'$ , we define  $\mathbf{X} = [1, 0, -1]$  and  $\mathbf{Y} = [-1, 2, -1]$  as fixed axis in the hexagonal plane. Of course,  $\mathbf{Z} = \mathbf{X} \times \mathbf{Y} = [1, 1, 1]$  coincides with  $\mathbf{z}'$  and lies along  $\mathbf{P}$ . The three reference frames used in this paper are summarized in Table I.

The lowest-order anisotropy energy of BiFeO<sub>3</sub> was included in the “canonical” model:

$$\mathcal{H}_{K_1} = -K_1 \sum_i S_{iZ}^2. \quad (12)$$

The two next-order anisotropy terms consistent with the rhombohedral symmetry [35] of BiFeO<sub>3</sub> are

$$\mathcal{H}_{K_2} = -\frac{1}{2}K_2 \sum_i S_{iZ} \times \left\{ (S_{iX} + iS_{iY})^3 + (S_{iX} - iS_{iY})^3 \right\}, \quad (13)$$

$$\mathcal{H}_{K_3} = -\frac{1}{2}K_3 \sum_i \left\{ (S_{iX} + iS_{iY})^6 + (S_{iX} - iS_{iY})^6 \right\}. \quad (14)$$

Whereas  $K_1$  is of order  $l^2|J_1|$  in terms of the dimensionless spin-orbit coupling constant  $l$ ,  $K_2$  and  $K_3$  are of order  $l^3|J_1|$  and  $l^4|J_1|$ , respectively [36,37]. These three terms have classical energies

$$E_{K_1} = \langle \mathcal{H}_{K_1} \rangle = -S^2 K_1 \sum_i \cos^2 \theta_i, \quad (15)$$

$$E_{K_2} = \langle \mathcal{H}_{K_2} \rangle = -S^4 K_2 \sum_i \cos \theta_i \sin^3 \theta_i \cos 3\phi_i, \quad (16)$$

$$E_{K_3} = \langle \mathcal{H}_{K_3} \rangle = -S^6 K_3 \sum_i \sin^6 \theta_i \cos 6\phi_i, \quad (17)$$

where the spin

$$\langle \mathbf{S}_i \rangle = S \left\{ \cos \phi_i \sin \theta_i \mathbf{X} + \sin \phi_i \sin \theta_i \mathbf{Y} + \cos \theta_i \mathbf{Z} \right\} \quad (18)$$

is given in the fixed reference frame defined above. Other anisotropy energies  $S^2 K_1' \sum_i \sin^2 \theta_i \cos 2\phi_i$  and  $S^4 K_2' \sum_i \sin^4 \theta_i \cos 4\phi_i$  vanish for the  $R3c$  crystal structure of BiFeO<sub>3</sub> [35,38].

For the distorted cycloid of the “canonical” model, both  $E_{K_1}$  and  $E_{K_3}$  are nonzero. Because the cycloid is mirror symmetric about  $Z = 0$ , the summation in  $E_{K_2}$  vanishes. Therefore,  $E_{K_2}$  will distort the cycloid to produce an energy reduction of order  $-(K_2)^2/|J_1|$ . Since  $E_{K_2}/E_{K_3} \sim l^2 \ll 1$ ,  $E_{K_2}$  can be neglected as a source of three-fold symmetry breaking compared to  $E_{K_3}$ .

### IV. MAGNETIC FIELDS

In this section and the next, we neglect the effects of domain pinning. The behavior of the domain wavevectors in an external field is then completely determined by the model developed above. The effects of pinning will be examined in Section V.

For  $K_3 > 0$ ,  $E_{K_3}$  favors spins that lie along one of the three three-fold axis  $\phi_i = 0$  and  $\pm 2\pi/3$ . With this additional anisotropy, the wavevectors  $\mathbf{q}_k$  rotate away from the three-fold axis with increasing field when the field does not itself lie perpendicular to a three-fold axis.

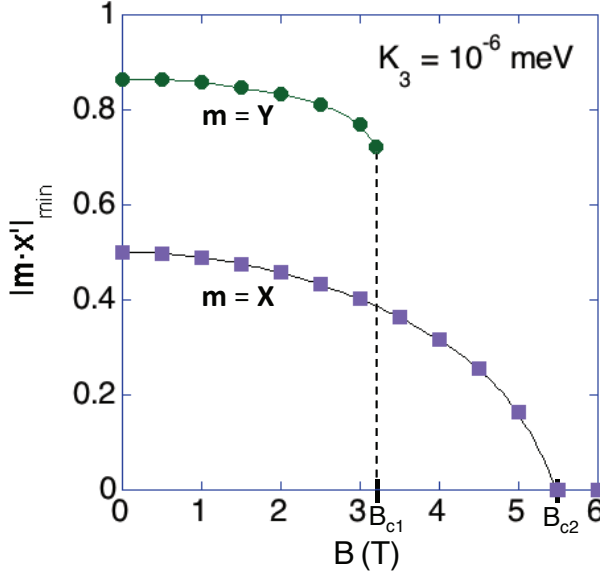


FIG. 3: (Color online) The wavevector  $|\mathbf{m} \cdot \mathbf{x}'|_{\min}$  for stable ( $\mathbf{m} = \mathbf{X}$ , squares) and metastable ( $\mathbf{m} = \mathbf{Y}$ , circles) domains versus field with  $K_3 = 10^{-6}$  meV.

#### A. Field along a three-fold axis or $\mathbf{m} = \mathbf{X}$

First take the field along a three-fold axis such as  $\mathbf{X}$  in Fig.1. Assuming that the system has been cooled from high temperature in zero field, all three domains with wavevectors  $\mathbf{q}_k$  will be equally occupied. But in large field, we expect that the stable domain will have wavevector  $\mathbf{q}'$  parallel to  $\mathbf{Y}$  and perpendicular to  $\mathbf{m}$ . For  $K_3 = 10^{-6}$  meV, the energy  $E = \langle \mathcal{H} \rangle$  is evaluated for several different wavevectors at each field. Defining  $E_0$  as the energy for  $K_3 = 0$  and  $B = 0$ , results for  $\Delta E = E - E_0$  are presented in Fig.2.

At zero field,  $\Delta E$  is minimized when  $\mathbf{x}'$  lies along a three-fold axis. Since  $\mathbf{m} = \mathbf{X}$  is itself a three-fold axis, minima appear when  $|\mathbf{m} \cdot \mathbf{x}'| = 1$  or  $1/2$ . With increasing field, the minimum at  $|\mathbf{m} \cdot \mathbf{x}'| = 1$  ( $\mathbf{x}' \parallel \mathbf{m}$ ) increases in energy so that this solution is only metastable. The stable solutions rotate from  $|\mathbf{m} \cdot \mathbf{x}'| = 1/2$  towards 0.

In addition to the critical field marking the transition into the CAF phase, we identify two lower critical fields. Below  $B_{c1} \approx 4.6$  T, the minima at  $|\mathbf{m} \cdot \mathbf{x}'| = 1$  survives so that the domain with wavevector along  $\mathbf{m}$  remains metastable. Above  $B_{c1}$ , that metastable domain disappears. As the field increases, the wavevectors of the stable domains rotate towards the orientation  $\mathbf{Y} \perp \mathbf{m}$ . In the absence of domain pinning, that rotation is complete at  $B_{c2} \approx 5.5$  T.

For each field, the dependence of energy on  $\mathbf{m} \cdot \mathbf{x}'$  can be described by a sixth order polynomial with even terms only. Based on the polynomial fits given by the solid curves in Fig.2, we obtain the minimum energy solutions for  $|\mathbf{m} \cdot \mathbf{x}'|$  at each field. We plot  $|\mathbf{m} \cdot \mathbf{x}'|_{\min}$  versus field in Fig.3. Above  $B_{c2} \approx 5.5$  T,  $\mathbf{q}$  lies perpendicular to  $\mathbf{m}$

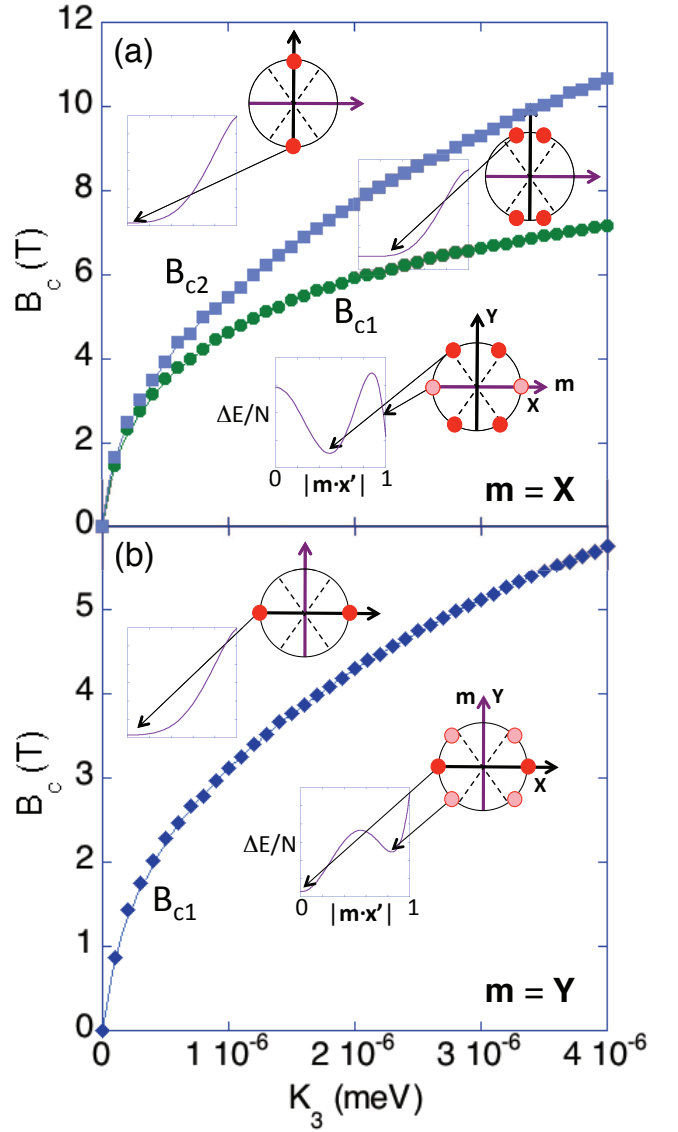


FIG. 4: (Color online) (a) The critical fields  $B_{c1}$  and  $B_{c2}$  versus three-fold anisotropy  $K_3$  for  $\mathbf{m} = \mathbf{X}$ . (b) The critical field  $B_{c1}$  versus  $K_3$  for  $\mathbf{m} = \mathbf{Y}$ . Insets schematically show the dependence of the satellite peaks on magnetic field and their energies.

and  $|\mathbf{m} \cdot \mathbf{x}'|_{\min} = 0$ .

The critical fields are plotted against the three-fold anisotropy  $K_3$  [39] in Fig.4(a). Both critical fields  $B_{c1}$  and  $B_{c2}$  and their difference  $B_{c2} - B_{c1}$  increase quite rapidly  $\sim \sqrt{K_3}$  for small  $K_3$ . We schematically sketch the dependence of the satellite peaks on magnetic field and their energies in the insets to Fig.4.

#### B. Field perpendicular to a three-fold axis or $\mathbf{m} = \mathbf{Y}$

When the field lies along  $\mathbf{Y}$ , the orientation of the stable domain does not change with field, i.e. domain 2 with



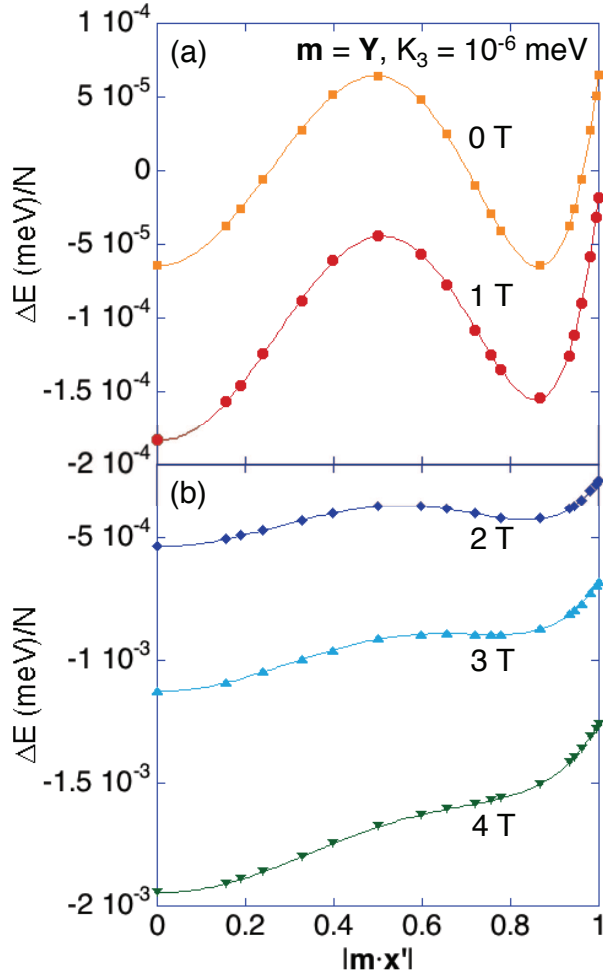


FIG. 5: (Color online) The energy  $\Delta E/N$  versus wavevector  $|\mathbf{m} \cdot \mathbf{x}'|$  for  $K_3 = 10^{-6}$  meV and field ranging from 0 to 4 T along  $\mathbf{Y}$ . Solid curves are fits to a sixth order polynomial in  $\mathbf{m} \cdot \mathbf{x}'$ .

$\mathbf{x}' = [1, 0, -1] \perp \mathbf{m}$  or  $\mathbf{m} \cdot \mathbf{x}' = 0$  is always stable. But as seen in Fig.5, domains 1 and 3 with  $\mathbf{x}' = [0, 1, -1]$  and  $[1, -1, 0]$  or  $|\mathbf{m} \cdot \mathbf{x}'| = \sqrt{3}/2$  are metastable for small fields and become unstable at high fields.

As in the previous subsection,  $\Delta E/N$  can be fit by a sixth order polynomial in  $\mathbf{m} \cdot \mathbf{x}'$  (even terms only). When  $K_3 = 10^{-6}$  meV, domains 1 and 3 are metastable below  $B_{c1} \approx 3.1$  T. With increasing field, the orientations  $\mathbf{x}'$  of the metastable domains rotate slightly towards the three-fold axis perpendicular to  $\mathbf{m}$ , as seen in the top curve of Fig.3. At  $B_{c1}$ ,  $|\mathbf{m} \cdot \mathbf{x}'| = \sqrt{2}/2$  so that the domain wavevectors  $\mathbf{q}$  have rotated from  $\theta = \pm\pi/6$  away from  $\mathbf{Y}$  at zero field to  $\pm\pi/4$  away from  $\mathbf{Y}$  at  $B_{c1}$  (although pinning will change this conclusion). Because the wavevector for domain 2 is already perpendicular to  $\mathbf{m}$  at zero field,  $B_{c2} = 0$ .

The dependence of  $B_{c1}$  on  $K_3$  is shown in Fig.4(b) [39]. Once again,  $B_{c1}$  scales like  $\sqrt{K_3}$  for small  $K_3$ .

## V. PINNING

The effects of pinning are essential to understand the behavior of the cycloidal domains in a magnetic field. Evidence for pinning was provided by recent SANS measurements [25]. For  $\mathbf{m} = \mathbf{X}$ , the wavevectors of the stable  $\mathbf{x}' = [1, -1, 0]$  and  $[0, -1, 1]$  domains remain unchanged up to about 5.5 T, above which they rotate towards  $\mathbf{Y} \perp \mathbf{m}$ . For  $\mathbf{m} = \mathbf{Y}$ , the wavevectors of the metastable  $\mathbf{x}' = [1, -1, 0]$  and  $[0, -1, 1]$  domains rotate towards  $\mathbf{X} \perp \mathbf{m}$  above about 5 T.

In a ferromagnet, domain pinning is caused by structural defects that locally change the exchange interactions and anisotropies, creating a complex energy landscape with barriers between different orientations of the magnetization  $\mathbf{M} = 2\mu_B \langle \mathbf{S}_i \rangle$  [40]. No doubt, these effects are also important in cycloidal spin systems. But the charge redistribution determined by the cycloidal wavevector  $\mathbf{q}$  may be even more important. Due to the strong magnetoelastic coupling in  $\text{BiFeO}_3$  [32,41], this charge redistribution is pinned by non-magnetic impurities. Although the total magnetoelastic energy is independent of  $\mathbf{q}$  (see Appendix A), the distortions  $\epsilon_{xx}$ ,  $\epsilon_{yy}$ , and  $\epsilon_{zz}$  of the rhombohedral structure separately depend on the wavevector orientation. In order to rotate  $\mathbf{q}$ , the magnetic field must drag this lattice distortion, pinned by non-magnetic impurities, through the crystal. Of course, this charge redistribution is absent in a collinear AF.

The susceptibility  $\chi_\perp$  for  $\mathbf{B} \parallel \mathbf{y}'$  or perpendicular to the plane of the cycloid is much larger than the susceptibility  $\chi_\parallel$  for  $\mathbf{B}$  within the cycloidal plane [19]. So the induced magnetization is  $\mathbf{M} = \chi_\perp \mathbf{B}_\perp$  where  $\mathbf{B}_\perp$  is the component of  $\mathbf{B}$  along  $\mathbf{y}'$ . The external field  $\mathbf{B} = \mathbf{B}_\perp + \mathbf{B}_\parallel$  plays two roles:  $\mathbf{B}_\perp$  produces the perpendicular magnetization  $\mathbf{M}$  and  $\mathbf{B}_\parallel$  exerts a torque  $\boldsymbol{\tau} = \mathbf{M} \times \mathbf{B}$  on  $\mathbf{M}$ .

### A. Microscopic model

To connect these considerations with our microscopic model, Fig.6 replots  $\Delta E/N$  versus  $\psi = \cos^{-1}(\mathbf{m} \cdot \mathbf{x}')$  while setting  $\Delta E/N = 0$  at  $\mathbf{m} \cdot \mathbf{x}' = 0$  or  $\psi = \pi/2$ . Using the angle definitions in Fig.7, note that  $\psi = \theta$  for  $\mathbf{m} = \mathbf{X}$  and  $\psi = \phi = \pi/2 - \theta$  for  $\mathbf{m} = \mathbf{Y}$ . We propose that a domain is pinned until the downward slope  $\varepsilon = -d(\Delta E/N)/d\psi$  exceeds the threshold  $\varepsilon_{\text{pin}} > 0$ . For  $\mathbf{m} = \mathbf{X}$ ,  $\varepsilon$  decreases with  $\psi$  in the neighborhood of  $\psi = \pi/3$ , as seen in the inset to Fig.6(a). As  $\psi$  increases, larger fields are required to fulfill the condition  $\varepsilon > \varepsilon_{\text{pin}}$ . A similar result is found for  $\mathbf{m} = \mathbf{Y}$  near  $\psi = \pi/6$ , as seen in Fig.6(b). In both cases,  $\psi$  satisfies the depinning condition  $\varepsilon = \varepsilon_{\text{pin}}$  as the field increases.

In the limit of strong pinning, the condition  $\varepsilon = \varepsilon_{\text{pin}}$  can be solved exactly. For large fields, the anisotropy can be ignored and  $\Delta E/N = -MB \sin \psi$ . To linear order in the field,  $M = \chi_\perp B \sin \psi$  so that  $\Delta E/N = -\chi_\perp B^2 \sin^2 \psi$  and  $\varepsilon = \chi_\perp B^2 \sin 2\psi$ .

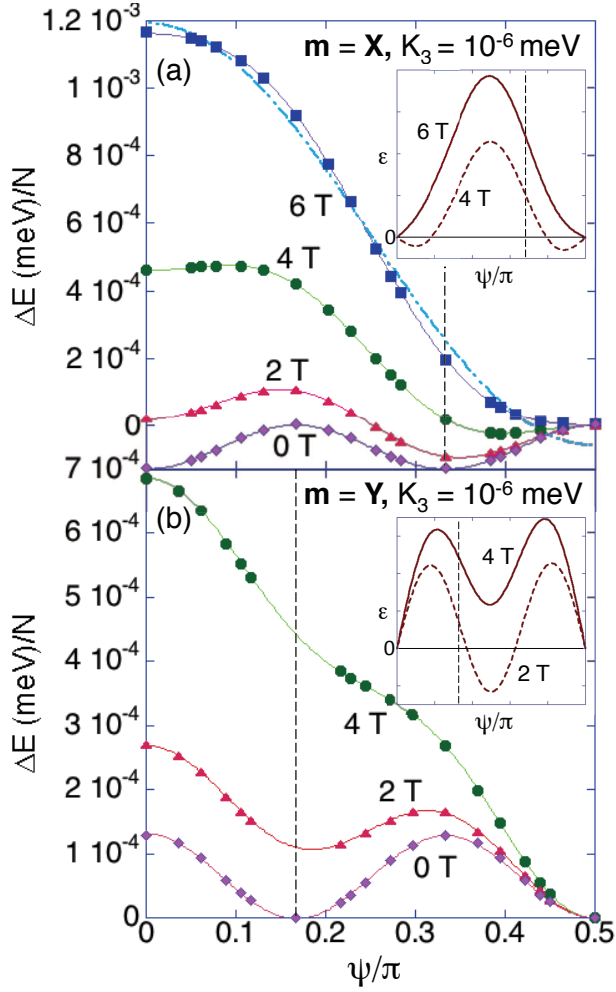


FIG. 6: (Color online) The energy  $\Delta E/N$  versus angle  $\psi = \cos^{-1}(\mathbf{m} \cdot \mathbf{x}')$  with  $\Delta E$  set to zero at  $\mathbf{m} \cdot \mathbf{x}' = 0$  or  $\psi = \pi/2$  for (a)  $\mathbf{m} = \mathbf{X}$  and (b)  $\mathbf{m} = \mathbf{Y}$ , both with  $K_3 = 10^{-6}$  meV. Dashed vertical lines are at  $\psi = \pi/3$  and  $\pi/6$ , respectively. Insets plot the derivative  $\varepsilon$  versus  $\psi$ . The dash-dot curve in (a) is a fit of the energy  $\Delta E/N$  to second order in  $\mathbf{m} \cdot \mathbf{x}' = \cos \psi$ .

For  $\mathbf{m} = \mathbf{X}$ , the energy  $\Delta E/N$  is fairly well-described by the form given above  $\propto \sin^2 \psi$  at high fields, as seen by the dash-dot curve in Fig.6(a) for 6 T. This agreement improves with increasing field. Consequently,  $\varepsilon \propto \sin 2\psi$  is close to the form in the inset to Fig.6(a) near  $\psi = \theta = \pi/3$  or  $\phi = \pi/6$ . So for strong pinning,  $\phi$  satisfies the condition

$$\sin 2\phi = \frac{\sqrt{3}}{2} \left( \frac{B_{\text{pin}}}{B} \right)^2, \quad (19)$$

where the pinning field  $B_{\text{pin}}^2 = 2\varepsilon_{\text{pin}}/\sqrt{3}\chi_{\perp}$  is defined so that  $\phi = \pi/6$  when  $B = B_{\text{pin}}$ .

For  $\mathbf{m} = \mathbf{Y}$ , the expression  $\varepsilon \propto \sin 2\psi$  is not satisfied until fields far above  $B_{c1}$ . So the simplified expression of Eq.(19) cannot be applied when  $\mathbf{m} = \mathbf{Y}$  and  $\psi = \phi = \pi/6$ . Hence, the depinning condition  $\varepsilon = \varepsilon_{\text{pin}}$  must be

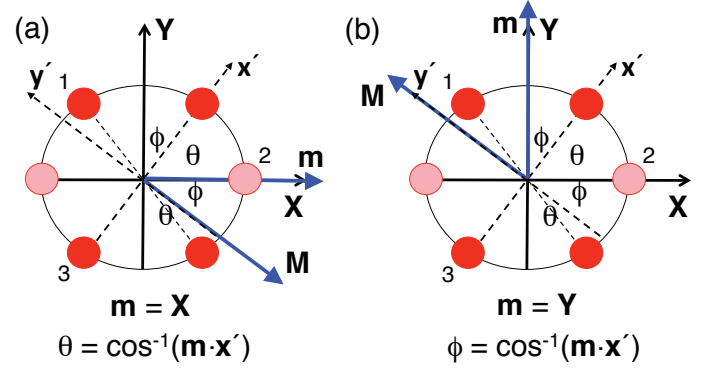


FIG. 7: (Color online) The magnetization and the cycloidal  $\mathbf{x}'$  and  $\mathbf{y}'$  axis for domain 3 with (a)  $\mathbf{m} = \mathbf{X}$  and (b)  $\mathbf{m} = \mathbf{Y}$ . Also shown are angles  $\theta$  and  $\phi = \pi/2 - \theta$ .

solved numerically.

Nonetheless, we can still draw some qualitative conclusions. The inset to Fig.6(b) indicates that domains 1 and 3 become unstable when  $\varepsilon > 0$  for  $\psi = \phi = \pi/4$ . So in the absence of pinning,  $\psi$  will grow from  $\pi/6$  in zero field to  $\pi/4$  at  $B_{c1}$ , in agreement with Fig.3. Taking pinning into account, there are two possible ways for domains 1 and 3 to evolve with field. If  $B_{\text{pin}} > B_{c1}$ , then the domains will disappear only after becoming depinned at  $B_{\text{pin}}$  with  $\psi = \pi/6$ . If  $B_{\text{pin}} < B_{c1}$ , then  $\psi$  will start rotating from  $\pi/6$  towards  $\pi/4$  above  $B_{\text{pin}}$  and stop rotating at  $B_{c1}$  with  $\psi < \pi/4$ . So the rotation towards  $\pi/4$  is not completed.

## B. Landau-Lifshitz equation

Another way to approach pinning is through the Landau-Lifshitz (LL) equation [42] for the time dependence of the magnetization:

$$\frac{\partial \mathbf{M}}{\partial t} = -2\mu_B \boldsymbol{\tau} + 2\alpha \mu_B \mathbf{M} \times \boldsymbol{\tau}, \quad (20)$$

where  $\boldsymbol{\tau} = \mathbf{M} \times \mathbf{B}_{\text{eff}}$  is the torque and  $\mathbf{B}_{\text{eff}}$  is an effective field that includes the effect of anisotropy. The first term in the LL equation produces the precession of  $\mathbf{M}$  about  $\mathbf{B}_{\text{eff}}$  and the second term gives the damping of  $\mathbf{M}$  as it approaches equilibrium. So the dimensionless parameter  $\alpha$  is proportional to the inverse of the relaxation time of the spins.

In the strong-pinning limit (see the discussion at the end of this section), we can neglect anisotropy and set  $\mathbf{B}_{\text{eff}} = \mathbf{B}$ . Since  $\mathbf{M}$  rotates within the  $\mathbf{X} - \mathbf{Y}$  plane, it can be written  $\mathbf{M} = M(\cos \varphi \mathbf{X} + \sin \varphi \mathbf{Y})$  so that

$$\begin{aligned} \boldsymbol{\tau} = \mathbf{M} \times \mathbf{B} = M \Big\{ & B_Z (\sin \varphi \mathbf{X} - \cos \varphi \mathbf{Y}) \\ & + (B_Y \cos \varphi - B_X \sin \varphi) \mathbf{Z} \Big\}. \end{aligned} \quad (21)$$



The LL equation then gives

$$\begin{aligned} \frac{d\varphi}{dt} &= -2\mu_B B_Z - 2\alpha\mu_B M (B_Y \cos \varphi - B_X \sin \varphi) \\ &= -2\mu_B B_Z - 2\alpha\mu_B \tau_Z. \end{aligned} \quad (22)$$

When  $\mathbf{m} = \mathbf{X}$ ,  $\varphi = -\phi$  and  $M = \chi_\perp B \cos \phi$  so

$$\tau_Z = \frac{\chi_\perp B^2}{2} \sin 2\phi = \frac{\varepsilon}{2}. \quad (23)$$

Hence, the torque along  $\mathbf{Z}$  and the energy derivative  $\varepsilon$  are simply related in the strong-pinning limit. Ignoring the precession of  $\mathbf{M}$  about  $\mathbf{Z}$  induced by  $B_Z$ , the relaxation of  $\phi$  towards equilibrium within the  $\mathbf{X} - \mathbf{Y}$  plane is determined by  $\tau_Z = \varepsilon/2$ .

Pinning in a ferromagnet is described by an effective field [26,27] that opposes the applied field, both along  $\mathbf{M}$ . In the strong-pinning limit of a cycloidal spin system, the external torque  $\tau$  along  $\mathbf{Z}$  is opposed by a pinning torque with maximum magnitude  $\tau_{\text{pin}}$ . The conditions  $\tau_Z = \tau_{\text{pin}}$  and  $\varepsilon = \varepsilon_{\text{pin}}$  are then equivalent. In terms of  $\tau_{\text{pin}}$ , the pinning field is given by  $B_{\text{pin}}^2 = 4\tau_{\text{pin}}/\sqrt{3}\chi_\perp$ .

For  $\mathbf{m} = \mathbf{X}$ , Eq.(19) is solved for  $\phi$  as a function of  $B/B_{\text{pin}}$  in Fig.8. As shown in the next section, Eq.(19) can be refined by expanding  $\chi_\perp$  in powers of  $B^2 \cos^2 \phi$ . Since  $\phi$  never reaches 0,  $B_{c2} = \infty$ .

Because the charge redistribution evaluated in Appendix A only depends on the direction of  $\mathbf{q}$ ,  $\tau_Z$  does not depend on the interior details of the cycloid such as its period or higher harmonics, but only on the magnetization  $\mathbf{M}$  induced by  $\mathbf{B}_\perp$ .

For  $\mathbf{m} = \mathbf{X}$ , experiments [25] observe pinning when  $B$  is lowered from  $B_{\text{pin}}$  but not as it is raised. This can be easily explained based on our model. For a fixed slope  $\varepsilon = \varepsilon_{\text{pin}}$ ,  $\Delta E/N \propto \cos^2 \theta / \sin 2\theta$  decreases with increasing  $\theta \geq \pi/3$ , as shown in the inset to Fig.8. So when  $B$  is raised from  $B_{\text{pin}}$ ,  $\phi = \pi/2 - \theta$  relaxes to a smaller value with lower energy. But when  $B$  is lowered from  $B_{\text{pin}}$ ,  $\phi$  would have to take a larger value with *higher* energy to satisfy the condition  $\varepsilon = \varepsilon_{\text{pin}}$  or  $\tau_Z = \tau_{\text{pin}}$ . This process is energetically prohibited at low temperatures. The pinning of domains with decreasing field is shown by the dashed lines in Fig.8. When the field is ramped up again with this value of  $\phi$ ,  $\mathbf{q}$  will only start rotating towards smaller values of  $\phi$  when the condition given by Eq.(19) is reached at the solid curve.

## VI. RESULTS

Let's use these ideas to examine the experimental results for BiFeO<sub>3</sub>. We separately discuss the two cases for field along  $\mathbf{X}$  or  $\mathbf{Y}$  examined in Section IV.

First take  $\mathbf{m} = \mathbf{X}$  as in Section IV.A. Since domain 2 with  $\mathbf{q}_2 \parallel \mathbf{m}$  becomes unstable when  $B > B_{c1} \approx 7$  T [25], the dependence of  $B_{c1}$  on  $K_3$  from Fig.4 implies that  $K_3 \approx 3.65 \times 10^{-6}$  meV. The pinning field  $B_{\text{pin}} \approx 5.6$  T for domains 1 and 3 is estimated from the comparison

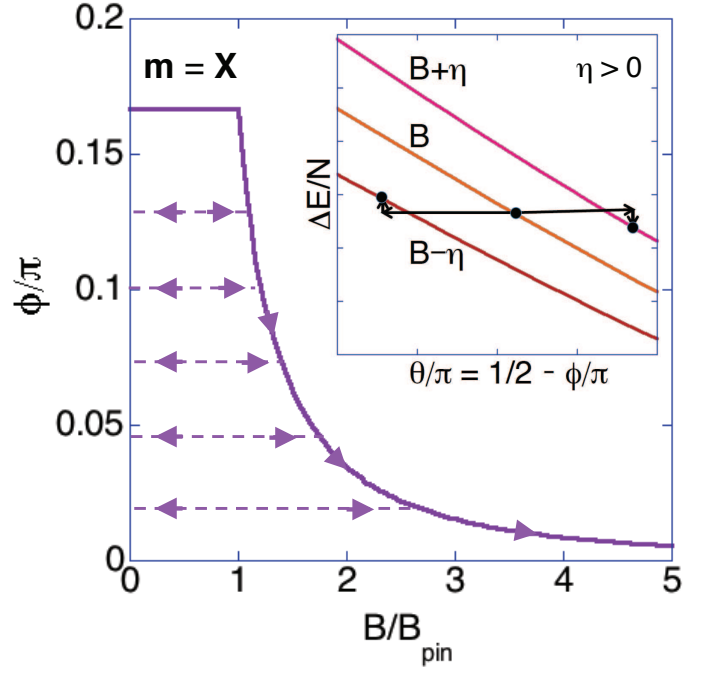


FIG. 8: (Color online) With  $\mathbf{m} = \mathbf{X}$ , the evolution of the rotation angle  $\phi$  for domains 1 and 3 with field  $B$  normalized by  $B_{\text{pin}}$ . The solid curve is for increasing field, the dashed lines are for decreasing or increasing field. Inset shows that  $\Delta E/N$  would have to rise with decreasing field to keep its downward slope  $\varepsilon$  unchanged. For increasing field,  $\Delta E/N$  drops.

between experimental results and Eq.(19) shown in the solid curve of Fig.9. Experimental data points are averaged over the four rotating satellites associated with domains 1 and 3. Measurements by Bordacs *et al.* [25] confirm that the wavevectors of domains 1 and 3 never become fully perpendicular to  $\mathbf{m} = \mathbf{X}$  but that  $\phi \rightarrow 0$  or  $\theta \rightarrow \pi/2$  with increasing field.

This model can be further refined by expanding  $\chi_\perp$  in powers of  $B^2 \cos^2 \phi$ . With  $b = B/B_{\text{pin}}$ ,

$$\chi_\perp(b) = \chi_0 + \chi_2 b^2 \cos^2 \phi, \quad (24)$$

where  $\chi_2$  is the non-linear susceptibility. This non-linear term includes the effects of higher harmonics [43] on the induced magnetization. Defining  $\gamma = \chi_2/\chi_0$ , we numerically solve

$$b^2 \left\{ 1 + \gamma b^2 \cos^2 \phi \right\} \sin 2\phi = \frac{\sqrt{3}}{8} (4 + 3\gamma) \quad (25)$$

with pinning field

$$B_{\text{pin}}^2 = \frac{4}{\sqrt{3}} \frac{\tau_{\text{pin}}}{\chi_0 + 3\chi_2/4}. \quad (26)$$

The solutions to Eq.(25) with  $\gamma = 0.2$  and  $0.4$  are plotted in the dashed curves of Fig.9. Evidently, a small value for  $\gamma$  further improves agreement with the measurements.

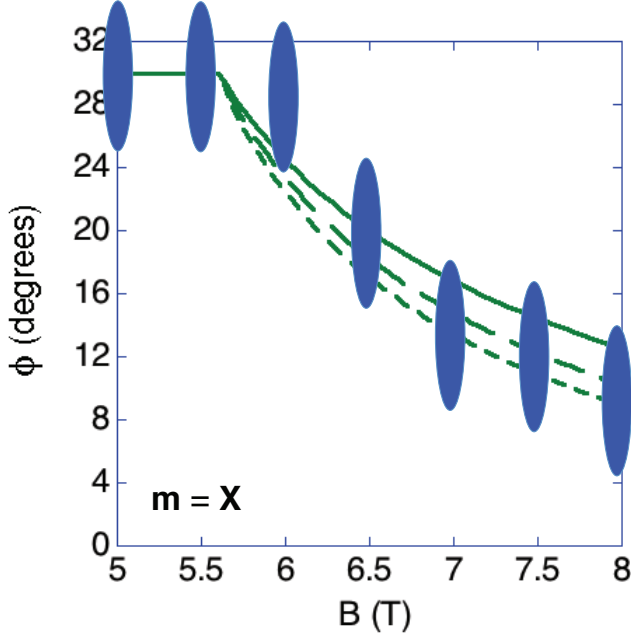


FIG. 9: (Color online) A comparison of model predictions with experimental data [25]. We set  $B_{\text{pin}} = 5.6$  T and  $\gamma = 0$  (solid), 0.2 (long dash), or 0.4 (short dash).

As mentioned above, the dependence of  $\phi$  on field given by Eq.(25) may only be approached at large fields if the condition for the strong-pinning limit is not met at  $B_{\text{pin}}$ .

Now take  $\mathbf{m} = \mathbf{Y}$  as in Section IV.B. Then domain 2 with  $\mathbf{q}_2 \perp \mathbf{m}$  is always stable. Unfortunately, the rotation of domains 1 and 3 cannot be treated in the strong-pinning limit. In particular,  $B_{\text{pin}}$  may differ from the earlier result for  $\mathbf{m} = \mathbf{X}$ . Experiments with  $\mathbf{m} = \mathbf{Y}$  indicate that domains 1 and 3 rotate by about 9 degrees before disappearing between 6 and 7 T. This implies that the second scenario discussed above with  $B_{c1} > B_{\text{pin}}$  is applicable. As expected,  $\psi = \phi$  has only increased to about  $39^\circ < 45^\circ$  at  $B_{c1}$ . But for  $K_3 = 3.65 \times 10^{-6}$  meV, Fig.4(b) predicts that  $B_{c1} \approx 5.6$  T, which is below the range of observed values where the domains disappear. This discrepancy can possibly be explained by a slight misalignment of the field out of the  $\mathbf{X} - \mathbf{Y}$  plane.

Since  $\tau_{\text{pin}}$  depends on the concentration and distribution of non-magnetic impurities, it may also change in different samples. Because the samples used in the spectroscopy [16] and SANS measurements [25] come from different sources, their pinning torques may be different as well. Based on the relative purities of these two samples,  $B_{\text{pin}}$  may be larger than indicated above for the sample used in the spectroscopy measurements. The observed spread [25] in wavevectors  $\mathbf{q}_1$  and  $\mathbf{q}_3$  near  $\mathbf{Y}$  for  $\mathbf{m} = \mathbf{X}$  also suggests that the pinning torque  $\tau_{\text{pin}}$  varies from one domain to another throughout the sample used in the SANS measurements. Unlike  $\tau_{\text{pin}}$ ,  $B_{c1}$  is determined by the relative energies of different domain wavevectors  $\mathbf{q}$  and is independent of sample quality.

The “strong-pinning” limit is reached when the anisotropy  $K_3$  makes a negligible contribution to the energy compared to the pinning field  $B_{\text{pin}}$ . Considering the contributions of the magnetic field and the anisotropy to the energy, the strong-pinning limit requires that  $B_{\text{pin}} M \gg 2S^6 K_3$  or

$$B_{\text{pin}}^2 \gg \frac{2S^6 K_3}{\chi_{\perp}}. \quad (27)$$

Using the definition of  $B_{\text{pin}}$  in terms of the pinning torque  $\tau_{\text{pin}}$ , the strong-pinning limit requires that  $\tau_{\text{pin}} \gg \sqrt{3}S^6 K_3/2$ . Taking  $K_3 = 3.65 \times 10^{-6}$  meV and  $\chi_{\perp}$  from measurements [3], these conditions require that  $B_{\text{pin}} \gg 2.5$  T and  $\tau_{\text{pin}} \gg 0.77 \mu\text{eV}$ .

Even for small  $B_{\text{pin}}$ , the strong-pinning limit can be reached when the field is sufficiently large that it dominates the energy. This condition is given by Eq.(27) with  $B$  replacing  $B_{\text{pin}}$ . So independent of  $B_{\text{pin}}$ , Eq.(19) for the dependence of  $\phi$  on field is approached when  $B \gg 2.5$  T.

## VII. DISCUSSION

This work resolves all of the discrepancies with earlier measurements listed in the introduction. Not too close to the poles at  $\pm \mathbf{z}'$ , the critical field  $B_{c3} > 16$  T above which the CAF phase becomes stable does not sensitively depend on the azimuthal angle  $\zeta$  because  $\mathbf{q}$  is then nearly perpendicular to  $\mathbf{m}$ . This explains the earlier discrepancy with measurements by Tokunaga *et al.* [23]. For any  $\zeta$ , Ref.[22] then predicts that  $B_{c3}$  will increase monotonically as the polar angle  $\vartheta$  decreases from  $\pi/2$  at the equator to zero at the poles.

Because it couples to the wavevector orientation but not to the individual spins, the strain does not directly affect the spin dynamics. However, it may be necessary to slightly raise  $K_1$  to compensate for the effect of  $K_3$ , which favors the spins lying in the  $\mathbf{X} - \mathbf{Y}$  plane [44]. Since the total magnetoelastic energy is independent of  $\mathbf{q}$ , it does not alter the relative energies of different wavevectors in Figs.2 and 5. Measurements of the lattice strain in a magnetic field along a three-fold axis like  $\mathbf{X}$  can test this hypothesis.

How does our estimate for  $K_3$  in BiFeO<sub>3</sub> compare with that in other materials? The constant  $K_3$  can be estimated from the angular dependence of the basal-plane magnetization or the torque. For  $\text{Co}_2\text{Y}$  ( $\text{Y} = \text{Ba}_2\text{Fe}_{12}\text{O}_{22}$ ) and  $\text{Co}_2\text{Z}$  ( $\text{Z} = \text{Ba}_3\text{Fe}_{24}\text{O}_{41}$ ),  $\tilde{K}_3 \equiv S^6 K_3/V_c \approx 600$  erg/cm<sup>3</sup> and 1500 erg/cm<sup>3</sup>, respectively [45] ( $V_c$  is the volume for one magnetic ion). For pure Co,  $\tilde{K}_3 \approx 1.2 \times 10^5$  erg/cm<sup>3</sup> [46]. Anisotropy energies are much larger for the rare earths than for transition-metal oxides [47]. While  $\tilde{K}_3 \approx 6300$  erg/cm<sup>3</sup> for Gd, it is about 1000 times higher for the heavier rare earths Tb, Dy, Ho, Er, and Tm. By comparison,  $K_3 = 3.65 \times 10^{-6}$  meV for BiFeO<sub>3</sub> corresponds to  $\tilde{K}_3 = 2.4 \times 10^4$  erg/cm<sup>3</sup>, about 4 times larger than for Gd but smaller than for pure Co.

Our discussion of domain pinning was motivated by previous results for ferromagnets. For a ferromagnet, thermally activated creep [48] allows the domain walls to move even when  $B < B_{\text{pin}}$ . It seems likely that a similar effect in BiFeO<sub>3</sub> allows the domains to rotate at nonzero temperature even when  $\tau_Z < \tau_{\text{pin}}$  or  $\varepsilon < \varepsilon_{\text{pin}}$ . Another interesting question is how the domain pinning depends on the rate of change of the magnetic field.

We conclude that the “canonical” model of BiFeO<sub>3</sub> must be augmented by three-fold anisotropy and magnetoelastic energies in order to explain the field evolution of a domain when  $\mathbf{q}$  is not perpendicular to  $\mathbf{m}$ . Over the past decade, our understanding of BiFeO<sub>3</sub> has greatly increased but so have the number of new mysteries to be solved. At least at low temperatures, we believe that the modified Hamiltonian presented in this work can be used to study the manipulation of multiferroic domains by magnetic and electric fields.

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## Appendix A: Magnetoelastic Coupling

This appendix describes the effects of the magnetoelastic coupling in BiFeO<sub>3</sub>. The magnetoelastic energy is given by

$$\begin{aligned} \frac{1}{V}H_{me} = & \frac{1}{2}c_{11}(\epsilon_{xx}^2 + \epsilon_{yy}^2 + \epsilon_{zz}^2) \\ & + c_{12}(\epsilon_{xx}\epsilon_{yy} + \epsilon_{yy}\epsilon_{zz} + \epsilon_{zz}\epsilon_{xx}) \\ & + \frac{g}{N} \sum_i \left\{ \epsilon_{xx}S_{ix}^2 + \epsilon_{yy}S_{iy}^2 + \epsilon_{zz}S_{iz}^2 \right\}, \quad (\text{A1}) \end{aligned}$$

where  $c_{11}$  and  $c_{12}$  are the elastic coupling constants,  $g$  is the magnetoelastic coupling strength, and  $\epsilon_{ii}$  are the strain components.

Minimizing this energy with respect to the strain components yields

$$\epsilon_{xx} = -\frac{g}{F} \left\{ (c_{11} + c_{12})M_{2x} - c_{12}(M_{2y} + M_{2z}) \right\}, \quad (\text{A2})$$

$$\epsilon_{yy} = -\frac{g}{F} \left\{ (c_{11} + c_{12})M_{2y} - c_{12}(M_{2x} + M_{2z}) \right\}, \quad (\text{A3})$$

$$\epsilon_{zz} = -\frac{g}{F} \left\{ (c_{11} + c_{12})M_{2z} - c_{12}(M_{2x} + M_{2y}) \right\}, \quad (\text{A4})$$

where

$$M_{2\alpha} = \frac{1}{N} \sum_i \langle S_{i\alpha}^2 \rangle \quad (\text{A5})$$

and  $F = (c_{11} + 2c_{12})(c_{11} - c_{12})$ . In terms of these variables, the magnetoelastic energy  $E_{me} = \langle H_{me} \rangle$  is given by

$$\begin{aligned} \frac{1}{N}E_{me} = & -g^2 \frac{V}{2N} \frac{c_{11} + c_{12}}{F} \left\{ M_{2x}^2 + M_{2y}^2 + M_{2z}^2 \right\} \\ & + g^2 \frac{V}{N} \frac{c_{12}}{F} \left\{ M_{2x}M_{2y} + M_{2x}M_{2z} + M_{2y}M_{2z} \right\}, \quad (\text{A6}) \end{aligned}$$

where  $V/N = a^3$ .

Transforming  $M_{2\alpha}$  into the local reference frame of the cycloid, we find

$$M_{2\alpha} = \frac{q_\alpha^2}{|\mathbf{q}|^2} M_{2x'} + \frac{1}{3} M_{2z'}, \quad (\text{A7})$$

which uses  $M_{2y'} \approx 0$ . For weak anisotropy,  $M_{2x'} \approx M_{2z'} = S^2/2$  and

$$\frac{1}{N}E_{me} = -\frac{g^2 S^4}{4F} \frac{V}{N} (3c_{11} - 2c_{12}), \quad (\text{A8})$$

which is independent of  $\mathbf{q}$ .

However, the individual strain components

$$\epsilon_{\alpha\alpha} = -\frac{gS^2}{2F} \left\{ \frac{q_\alpha^2}{|\mathbf{q}|^2} (c_{11} + 2c_{12}) + \frac{1}{3} (c_{11} - 4c_{12}) \right\} \quad (\text{A9})$$

do depend on  $\mathbf{q}$ . In fact, roughly 75% of the strain depends on the wavevector orientation. Impurities clamp this lattice strain within the sample. Because the magnetic field must drag this distortion while rotating the cycloidal wavevector  $\mathbf{q}$ , impurities pin the orientation of the wavevector at low fields. Note that the volume change

$$\frac{\Delta V}{V} = \epsilon_{xx} + \epsilon_{yy} + \epsilon_{zz} = -\frac{gS^2}{c_{11} + 2c_{12}} \quad (\text{A10})$$

is independent of the wavevector orientation.

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- $$\frac{\mathcal{H}_{D_2}}{N} \rightarrow \frac{3D_2}{2\pi} \mathbf{z}' \cdot \int_0^{2\pi} d\rho \left( \mathbf{S}_\rho^{(1)} \times \mathbf{S}_\rho^{(2)} \right),$$
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