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# A phenomenological theory for polarization flop in spiral multiferroic $TbMnO_3$

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**Abstract.** A phenomenological Landau theory has been used to explain magnetic field-driven polarization flop in $TbMnO_3$. The Néel wall-like magnetic structure in spiral multiferroics induces a space-dependent internal magnetic field which exerts a torque on spins to rotate bc-spiral to ab-spiral. The external magnetic field is argued to be competing with easy axis anisotropy and the system stabilizes when anisotropy is minimum. With the help of Landau free energy with DM magnetoelectric coupling and a general ansatz for magnetization, the phenomenon of polarization flop has been explained. Relation between $T_{\rm flop}$ and critical magnetic field has been established and found to be in good agreement with the experiment. This could be an indication that anisotropy of the system is temperature- and magnetic field-dependent.

**Keywords.** Multiferroics; magneto-optical properties; spin density wave.

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Electric control of magnetization and magnetic control of polarization have been long sought phenomena for researchers because of their potential applications in spintronics and multiple state memory devices [1,2]. Magnetic control of polarization has been experimentally achieved by Kimura *et al* [3,4], where they reported not only the discovery of ferroelectricity in a perovskite manganite $TbMnO_3$ (where the effect of spin frustration causes ferroelectricity) but also the polarization flop of 90° with the application of magnetic field. $TbMnO_3$, which has an orthorhombically distorted perovskite structure, shows a sinusoidal incommensurate antiferromagnetic $Mn^{3+}$ spin ordering below 41 K. Below 28 K, it sets a commensurate bc-spiral magnetic structure, propagating along *b*-axis, associated with a polarization ($P_z$) along the *c*-axis. When an external magnetic field is applied along *b*-axis above a threshold field (4 T), the polarization along the *c*-axis decreases and a polarization along the *a*-axis ($P_x$) appears. At a critical field (say $H_{\rm c} = 8$ T at temperature 20 K), $P_z$ becomes zero and $P_x$ saturates to a maximum value. Soon after, association of polarization with magnetic structure was explained by spin current model

[5], or equivalently by inverse Dzyaloshinskii–Moriya interaction [6,7] where the electric polarization is expressed as

$$P = \sum_{i,j} A\hat{e}_{i,j} \times (\vec{S}_i \times \vec{S}_j).$$

Here $\hat{e}_{i,j}$ denotes the unit vector connecting the interacting neighbour spins $\vec{S}_i$ and $\vec{S}_j$ and $A$ is a constant relevant to the spin exchange interaction. In a recent study, Mochizuki and Furukawa [8] have taken a general microscopic Hamiltonian for spiral multiferroic, and using Monte-Carlo simulation, have reproduced $T$–$H_{\mathrm{ex}}$ phase diagram for $TbMnO_3$ and $DyMnO_3$. They claim that applied magnetic field reduces DM energy and a competition between DM interaction and other interactions results in polarization flop.

This paper proposes a simple way to explain the phenomenon of polarization flop which is a result of the flop of bc-spiral plane to ab-spiral plane [5] (figure 1 shows bc- and ab-spiral structures as in $TbMnO_3$). The flop occurs when a space-dependent magnetic field in the presence of DM interaction exerts a torque upon spins ($c$-axis) to rotate them in a perpendicular direction ($a$-axis). The system stabilizes at the balance of competing external magnetic field and easy axis anisotropy. An expression showing the relation between $T_{\mathrm{flop}}$ and $B_{\mathrm{flop}}$ has been derived which agrees with the experiment. In the present paper, we make an attempt to explain polarization flop using phenomenological Landau theory and argue that Néel wall-like structure and DM coupling are responsible for the same. The paper is arranged in the following manner: in the next section, some properties of Néel walls have been summerized. Further, the spatial profiles of magnetization and polarization in both a conventional 180° Néel wall and a spiral multiferroic have been shown. Then in the next section taking Landau free energy with DM magnetoelectric coupling and a general magnetic ansatz, the phenomenon of polarization flop is explained. The temperature dependence of polarization flop is studied and relationship between $T_{\mathrm{flop}}$ and $B_{\mathrm{flop}}$ is established and compared with the experiment. The discussions of the results is given at the end.

In general, magnetic domain walls are a result of competition between exchange and anisotropy in the system. The width of a domain wall is equal to $\pi\sqrt{A/w}$, where $A$ is the typical nearest-neighbour Heisenberg exchange stiffness constant and $w$ is the typical anisotropy constant [9]. Domain wall energy is given by $4\sqrt{Aw}$ showing the competition between exchange and anisotropy. In the case of soft or amorphous materials characterized by a vanishing anisotropy constant $w$, one uses the magnetostatic exchange length defined by $l_{\mathrm{ex}} = \sqrt{A/M_s^2}$ where $M_{\mathrm{s}}$ is the saturation magnetization. In all these cases, the wall width $\delta$ is obtained from the exchange length using the equation $\delta = \pi l_{\mathrm{ex}}$. In a regime where the thickness of a ferromagnetic film becomes comparable to the exchange length ($\sqrt{A/w}$), Néel-type walls are created. In the case of Néel walls, due to non-vanishing divergence of magnetization, an internal field is induced which has important physical

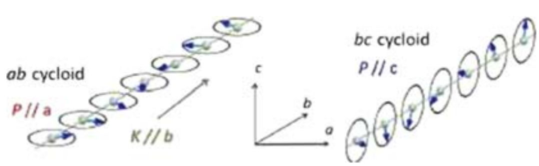


**Figure 1.** ab and bc cycloidal spiral structure as in the case of $TbMnO_3$.

and mathematical consequences. Unlike the Bloch wall where only two energy components (exchange and anisotropy) exist, in Néel wall an additional energy competes due to the internal energy. Thus, the mathematical description of Néel walls entails the introduction of an internal magnetic field $H$ formed by induced pole density $\rho$ resulting from non-vanishing divergence of $M$. Considering that magnetization is expressed as $\vec{M} = (0, M\cos(\theta(y)), M\sin(\theta(y)))$, we obtain induced pole density as

$$\rho = -\mathrm{div}\vec{M} = -\frac{\partial M_y}{\partial y} = -M_\mathrm{s}\frac{\mathrm{d}(\cos\theta)}{\mathrm{d}y}. \quad (1)$$

Internal field can be obtained from the induced pole density using general theorems of electromagnetism which comes out to be

$$H_\mathrm{int}(y) = \frac{1}{\pi}\int_{-\infty}^{\infty} \rho(y')\tan^{-1}\left[\frac{t}{2(y-y')}\right]\mathrm{d}y' \quad (2)$$

along $y$-direction. $t$ represents the thickness of the film. The total energy of Néel domain wall contains exchange, anisotropy and a Zeeman term, that is

$$F = M_\mathrm{s}\int_{-\infty}^{\infty} \frac{A}{2}\left(\frac{\partial\vec{M}}{\partial y}\right)^2 + \frac{w}{2}M_z^2 - M_y H(y)\mathrm{d}y. \quad (3)$$

On minimization with respect to $\theta$ we obtain a second-order nonlinear differential equation

$$2A\frac{\partial^2\theta}{\partial y^2} + w\sin(2\theta) + M_\mathrm{s}H(y)\sin\theta = 0. \quad (4)$$

Solution of this equation gives a 1D Néel domain wall. If an external magnetic field is applied along the $y$-axis, it just adds up with the internal field in Zeeman term and gives similar result qualitatively. Figure 2 shows spatial profile of $\theta$ for various magnetic fields. It shows that as magnetic field increases, the wall width ($\pi\sqrt{A/w}$) also increases which can be thought as decreasing the anisotropy $w$ of the system. This important relationship between anisotropy and magnetic field will be used in the next section to interpret polarization flop.

It has already been discussed in previous studies [10] that flexoelectric interactions are present in all ferromagnets and their presence results in non-zero polarization within a domain wall of Néel character [11]. In figure 3, the spatial profiles of the polarization and magnetization in conventional Néel wall are shown. The spatial profiles of magnetization and polarization in the bc-spiral of $TbMnO_3$ are plotted by considering flexoelectric-type magnetoelectric coupling (figure 4).

In the spiral magnetic structure of multiferroic $TbMnO_3$, due to Néel wall-like structure, an internal magnetic field is present which plays an important role in polarization flop in these systems. In $TbMnO_3$ bc-spiral and ab-spiral are two ground states but due to the $z$-axis anisotropy, bc-axis becomes the preferred ground state for this system. Thus, the flop of the bc-spiral to the ab-spiral can be considered as a 90° Bloch domain wall in magnetic field space where the two boundaries are 0 and $H_\mathrm{c}$. As external magnetic field increases, easy axis anisotropy decreases making ab-spiral as likely as bc-spiral. Also,

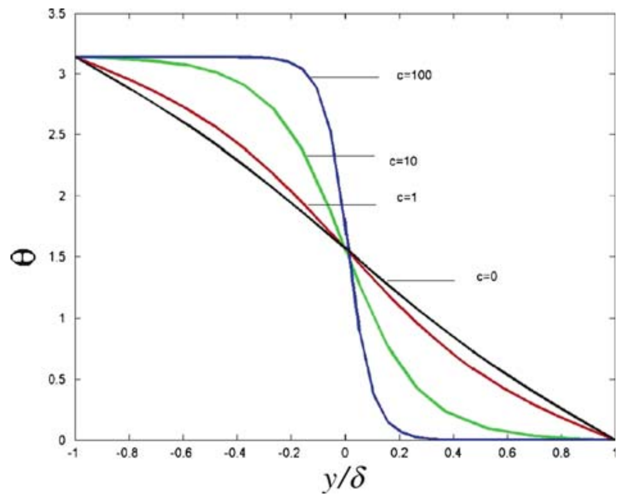


**Figure 2.** Change of $\theta$ with $y/\delta$ in Néel domain wall for four values of magnetic field ($c$ is the normalized magnetic field $= H_{\text{ext}}/H_w$). Figure shows that domain wall width increases with increase in magnetic field.

due to the presence of Dzyaloshinskii–Moriya interaction term, this magnetic field exerts a torque upon $M_z$ in $x$ direction. This also explains the presence of fluctuation of $P_x$ in $TbMnO_3$ at zero magnetic field, the sign of which reflects in the Raman spectra of $TbMnO_3$ with $E \| a$ [12].

We consider a Landau free energy for spiral magnetic structure multiferroic with inverse DM coupling as in previous studies [13].

$$F = \int f \mathrm{d}^3 r, \tag{5}$$

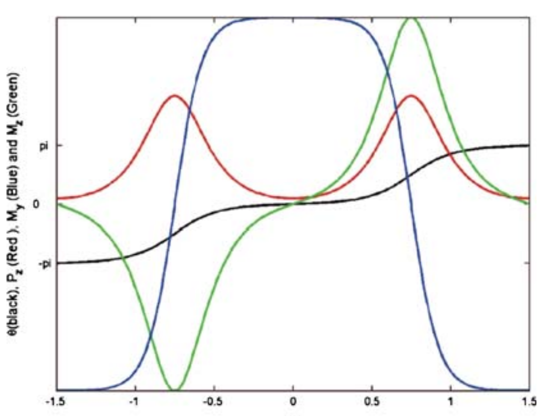


**Figure 3.** Spatial profile of magnetization and polarization over the complete wavelength of a Néel wall. Polarization is maximum when magnetization parallel to polarization is maximum.

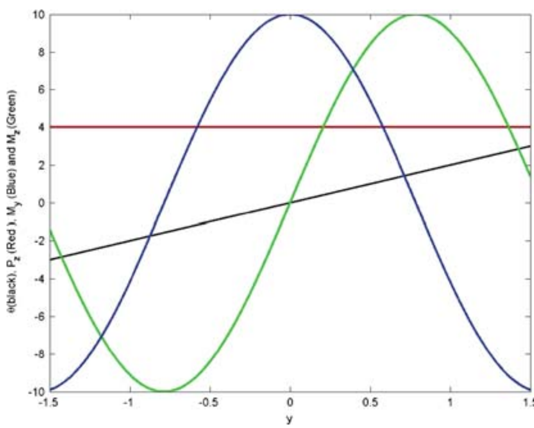


**Figure 4.** Spatial profile of polarization and magnetization over the complete wavelength of magnetic spiral structure with $\theta = ky$. Polarization is constant with space.

where $f$ is the free energy density given by

$$f = \frac{a}{2}\vec{M}^2 + \frac{u}{4}\vec{M}^4 + \frac{w}{2}M_z^2 + \frac{\gamma}{2}(|\nabla M|)^2 + \frac{\alpha}{2}(\vec{\nabla}^2\vec{M})^2 + \frac{b}{2}\vec{p}^2 + \eta\vec{p}\cdot(\vec{M}\times\vec{\nabla}\times\vec{M}) - \vec{M}\cdot\vec{H}. \quad (6)$$

Here, $a = a_0(T - T_c)$, $w$, $\alpha$, and $\eta$ are positive and $\gamma$ is negative. This, however, is not the most generalized Landau free energy for spiral magnetic structure multiferroic, but it has been written specifically for $TbMnO_3$. For properties like Raman spectra of $TbMnO_3$, in addition to the inverse DM, other forms of coupling have been found responsible [14,15]. For conical cycloidal multiferroic, e.g. $CoCr_2O_4$, due to the absence of geometric anisotropy, in free energy equation, uniaxial anisotropy term will be absent and spin-orbit coupling causes the surface term to be anisotropic for two axes [16]. Anisotropy plays a significant role in the formation of spiral magnetic structure. Therefore, in the case of geometrically isotropic systems (e.g. $CoCr_2O_4$) anisotropy appears by other means. Here, we have taken inverse DM coupling particularly due to the following reasons: (1) inverse DM coupling term is symmetrically allowed, (2) it can explain a number of properties of spiral multiferroic and (3) we believe it to be responsible for magnetic field-dependent polarization flop in $TbMnO_3$. We retain the lower-order terms of free energy (neglecting $\frac{u}{4}\vec{M}^4$ and $\frac{\alpha}{2}(\vec{\nabla}^2\vec{M})^2$ terms) and consider a general magnetic ansatz as

$$\vec{M} = M\sin(\theta(y))\sin(\phi\{H(y)\}\hat{e}_1 + M\cos(\theta(y))\hat{e}_2 + M\sin(\theta(y))\cos(\phi\{H(y)\}\hat{e}_3), \quad (7)$$

where $M$, the amplitude of magnetization, is taken to be the same for the three axes for the sake of simplicity and shows temperature dependence. $\theta$ is the space-dependent part of magnetization and for commensurate spiral phase, it is equal to $ky$. $\phi$ is magnetic field dependent part of the magnetization. Since magnetic field H(y) is space-dependent, $\phi$ is also space-dependent. Here magnetic field $H(y)$ comprises two parts, the external applied field $H$ and the internal induced field $H_{\text{int}}$ (space-dependent).

$$H(y) = H + H_{\text{int}}(y).$$

Now the two spiral structures at different magnetic fields can be obtained from this ansatz:

$$H = 0, \quad \phi\{H(y)\} = 0, \quad \vec{M} = (0, M_2\cos(\theta), M_3\sin(\theta))$$

$$H = H_c, \quad \phi\{H(y)\} = \pi/2, \quad \vec{M} = (M_1\sin(\theta), M_2\cos(\theta), 0). \tag{8}$$

Omitting higher order terms such as $(\frac{u}{2}M^4)$ and $\frac{\alpha}{2}(\vec{\nabla}^2\vec{M})^2$ in the free energy (eq. (7)) and putting ansatz (8) for the magnetic order parameter, we minimize the free energy density with respect to $p_x$ and $p_z$ to obtain space-dependent polarization along the $a$- and $c$-axis respectively,

$$p_x = -\frac{\eta M^2}{b}\left(\frac{\partial\theta}{\partial y}\right)\sin(\phi) - \frac{\eta M^2}{2b}\sin(2\theta)\left(\frac{\partial\phi}{\partial H}\right)\left(\frac{\partial H}{\partial y}\right)\cos(\phi), \tag{9}$$

$$p_z = -\frac{\eta M^2}{b}\left(\frac{\partial\theta}{\partial y}\right)\cos(\phi) + \frac{\eta M^2}{2b}\sin(2\theta)\left(\frac{\partial\phi}{\partial H}\right)\left(\frac{\partial H}{\partial y}\right)\sin(\phi). \tag{10}$$

Here, $\theta = ky$ and $H_{\text{int}}(y) \approx M_y = M\cos(ky)$. Putting eqs (11) and (12) in free energy and minimizing with respect to $\phi$, we obtain second-order non-linear differential equation in $\phi$ as

$$\frac{\partial^2\phi}{\partial H^2} - \xi\sin(2\phi) = 0, \tag{11}$$

where $\xi = (-2w/3\gamma k^2M^2) > 0$ is an important parameter which shows the competition between exchange $(\gamma)$ and anisotropy $(w)$ and the two decide the value of the critical magnetic field.

A trial solution to the above nonlinear differential equation which satisfies boundary conditions (10) is

$$\phi = 2 * \tan^{-1}(\exp(1 - 1/(\sqrt{\xi} * H)). \tag{12}$$

With the trial solution (14), the second-order nonlinear differential equation (13) has been solved using the multiple shooting method. The variation of $\phi$, with normalized magnetic field $(H/H_c, H_c = 1/(\sqrt{\xi}))$ is shown in figure 5. The magnetic field dependence of $\phi$ provides a way to look for polarization flop. In order to obtain net polarizations we integrate eqs (11) and (12) over $y$ and get the expressions for $P_x$ and $P_z$ as

$$P_x = \int_0^{2\pi/k} p_x\mathrm{d}y = -\frac{\eta M^2}{b}\int_0^{2\pi/k}\sin(\phi)\mathrm{d}y, \tag{13}$$

$$P_z = \int_0^{2\pi/k} p_z\mathrm{d}y = -\frac{\eta M^2}{b}\int_0^{2\pi/k}\cos(\phi). \tag{14}$$

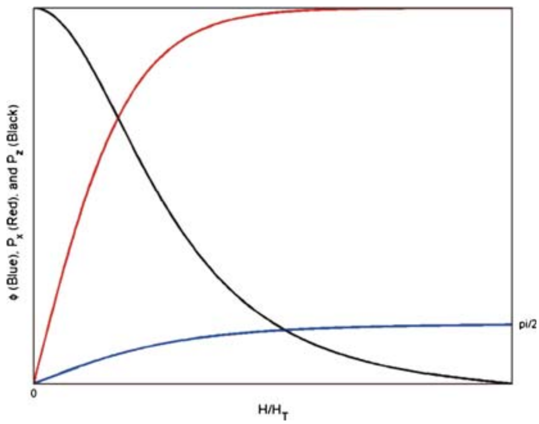


**Figure 5.** Flop of polarization from $P_z$ to $P_x$ on application of the magnetic filed.

Equations (13) and (14) have been integrated numerically over $y$ and show polarization flop as shown in figure 5. It has already been found experimentally by Kimura [3] that $H_c$ is temperature-dependent. To derive the relationship between $T_{flop}$ and $H_c$ we put the boundary condition in eq. (14) which is, $\phi = \pi/2$ for $H = H_c$ and $T = T_{flop}$. Thus, we have

$$H_c^2 = -\xi_0/(T_{flop} - T_l), \tag{15}$$

where $\xi_0 = -3\gamma K^2 M^2/2w_0$. In this relation the temperature dependence comes through temperature dependence of anisotropic term ($w = w_0(T - T_l)$) which is zero at and above $T_l$.

Fitting expression (15) on experimental data [3], $\xi_0$ comes out to be 500 $T^2K$ (figure 7). Using the value of $C_0$, polarizations $P_x$ and $P_z$ with normalized magnetic field ($H/H_c$) is plotted as shown in figure 5.

With this relation (15), we also obtain the temperature dependence of $\phi$. Now we plot the temperature dependence of polarizations $P_x$ and $P_z$ (figure 6) which is two-fold. First

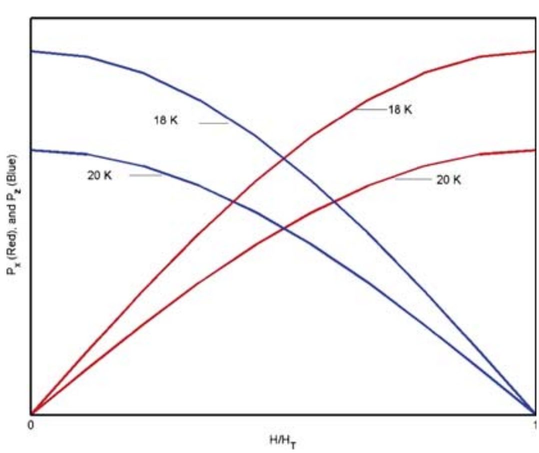


**Figure 6.** Flop of polarization at different temperatures.

one comes through temperature dependence of magnitude of $M_z/M_x$ below $T_l$ which increases the magnitude of polarization with decreasing temperature. An additional temperature dependence comes through the temperature dependence of $\phi$ which acts only in the presence of magnetic field and below $T_{\text{flop}}$ decreases $H_c$ with decreasing temperature (figure 7).

The solution of eq. (13) with the help of boundary value equations (10) is the same as that of the 90° Bloch domain wall where the wall width is $1/\sqrt{\xi}$, i.e. $\propto \sqrt{\gamma/w}$ and domain wall energy is proportional to $\sqrt{w\gamma}$. Here, if the anisotropy is temperature-dependent, the domain wall energy as well as domain wall width and hence boundaries of the domain wall become temperature-dependent.

Conclusively, the phenomenon of polarization flop can be explained as follows: Both the ab- and bc-spiral structures could be the possible ground states but due to the anisotropy along the $c$-axis in $TbMnO_3$ bc-spiral wins over ab-spiral. A Néel wall-like magnetic structure exhibits a space-dependent magnetic field which, as a consequence of DM interaction, exerts a torque on the c-spins to rotate them along the a-axis. Also, the increasing external magnetic field along the propagation vector decreases the anisotropy by overcoming the energy gap between ab- and bc-spirals. Thus, the torque competes with the exchange-to-anisotropy ratio and the system stabilizes at ab-spiral when anisotropy is minimum. Thus polarization flop due to the rotation of cycloidal plane from bc-spiral to ab-spiral can be regarded as the formation of a virtual 90° Bloch domain wall in the magnetic field space. The boundaries of this domain wall ($H_c$) is temperature-dependent due to temperature dependence of the uniaxial anisotropy. Thus, domain wall energy increases and domain wall width ($H_c$) decreases with decreasing temperature. When anisotropy is absent, no external magnetic field would be required for such polarization flop. However, there would be no formation of spiral structure in the absence of anisotropy. Even if our findings in the present work makes the anisotropic term in the free energy temperature dependent, the temperature dependence of the electromagnon and related properties will be unaltered [17]. As the spiral structure, polarization flop and $H$–$T$ phase diagram of $DyMnO_3$ are very similar to those of $TbMnO_3$, this theory may be applicable to polarization flop in $DyMnO_3$.

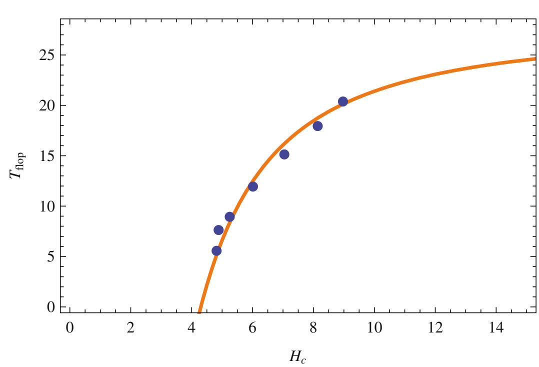


**Figure 7.** Relation between $T_{\text{flop}}$ and $H_c$ (experimental data are taken from ref. [1]). Experimentally the curve does not touch $x$-axis because at lower temperature (7 K) ordering of $Tb^{3+}$ ions takes place.

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