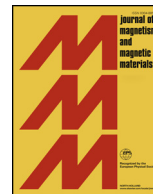




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Research articles

Novel type of spin cycloid in epitaxial bismuth ferrite films

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A B S T R A C T

We report on the novel cycloids that can be realized in epitaxial BiFeO₃ films. We found two featured (1 1 0) and (1 $\bar{1}$ 0) plane cycloids in the (1 1 0) oriented BiFeO₃ film and considered the action of compressive and tensile deformations on the areas of the cycloids stability. Our findings show that additional magnetic anisotropy induced by the strains allocates cycloids with the definite directions of spin rotation and electric polarization and change the properties of space modulated structures. Until now, it was believed that polarization is linked with antiferromagnetic order throughout the plane of spin rotation, namely it was considered that polarization always lies in the cycloid spin rotation plane. We show that due to the action of epitaxial strains spin rotational plane of the cycloid deviates from the plane containing intrinsic spontaneous polarization P_s .

1. Introduction

Bismuth ferrite (BiFeO₃) is currently in the focus of intensive scientific research, unique combination of ferroelectric and magnetic ordering established at room temperatures offers attracting promises for spintronics applications. A key issue in the BiFeO₃ – based devices is electric control of magnetic states sensitive to external influences. Various antiferromagnetic phases are revealed in thin BiFeO₃ films where non-collinear and collinear magnetic order can be engineered by epitaxial strains.

Due to the Dzyaloshinskii – Moriya interactions (DMI) supported by electric polarization in BiFeO₃-like multiferroics, a tendency to form incommensurate long periodical structures always exists in these compounds. The ground state of “proper multiferroics” whose magnetic and ferroelectric structures are formed independently [1] is incommensurate structure of cycloidal type. First, the spin cycloid was discovered in BiFeO₃ by I. Sosnowska et al. in 1982 [2]. The mechanism explaining origin of space modulated structures (SMS) and the solution describing cycloidal state was suggested in 1995 in Ref. [3]. Further on, it was established that spin cycloids optimize the coupling between antiferromagnetic and ferroelectric orders. As was shown in Refs. [4] electric field subsequently switches electric polarization and changes the orientation of spin rotation plane of the cycloid. That gives the key for practical implementation of the cycloids in spintronic devices. Complete electric control of magnetic states in the BiFeO₃ – based bias systems can be achieved via polarization switching accompanied with

the spin flop transitions between antiferromagnetic cycloids; in this case the knowledge of the correlation between polarization and the cycloidal state becomes crucial.

The properties of SMS in BiFeO₃ have been extensively explored in a number of theoretical and experimental works [3–12]. One of the main features of considered therein cycloids is the presence of polarization in their spin rotational plane.

However, the different situation can also exist. In our article, we report on the novel magnetic state: spin cycloid whose spin rotational plane deviates from the vector of spontaneous polarization. This situation can be realized in a film due to the action of epitaxial stresses affecting magnetoelectric properties of BiFeO₃. Strains (uniaxial, biaxial, hydrostatic ones) allocate prevailing components of electric polarization and can even totally suppress it [13–15]. Striction effects lead to appearance of additional magnetic anisotropy that influences ground magnetic state.

Herein, we explore properties of a featured antiferromagnetic cycloid in the (1 1 0) oriented BiFeO₃ film. We confirm the existence of the cycloids modulated along [0 0 1], 1 $\bar{1}$ 0 axes with (1 $\bar{1}$ 0), (1 1 0) spin rotational planes that do not contain polarization vector. We determine characteristics of space-modulated phase, find the range of material parameters required for existence of the cycloids and calculate the critical points of phase transitions.

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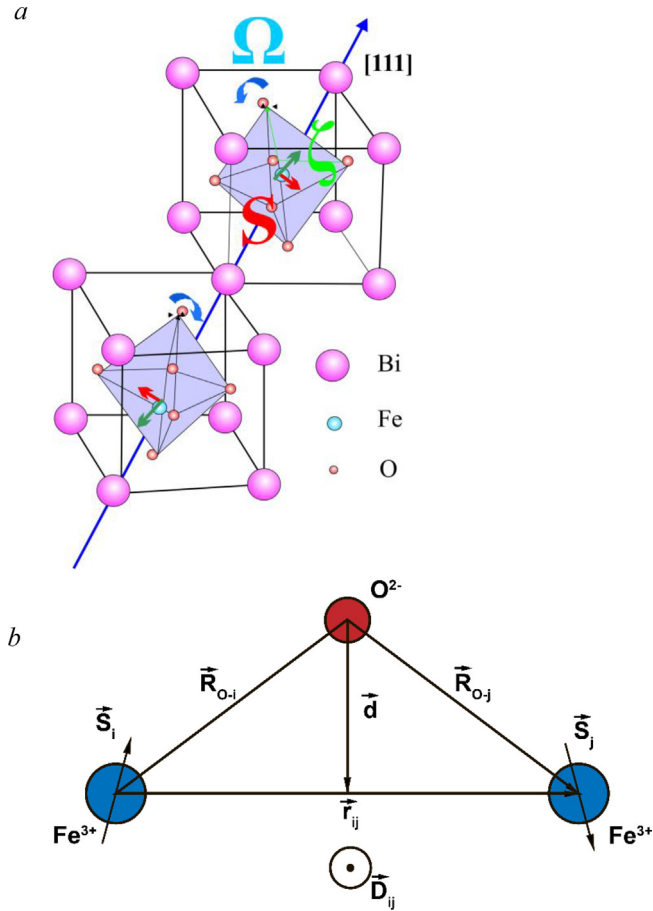


Fig. 1. a) Unit cell of BiFeO₃ (Ω is the vector of oxygen octahedrons antirotations, ζ is the displacement of Fe³⁺ and Bi³⁺ ions along the principal axis $[111]$, S is the spin of Fe³⁺ ion), b) illustration of Dzyaloshinskii – Moriya interaction (DMI) and the orientation of local DM vector.

2. Model and basic equations

The crystal structure of BiFeO₃ originates from ABO₃ perovskites, its unit cell is doubled and distorted ABO₃ unit cell formed due to three distortions: relative displacement of ions along principle crystal axis $\langle 111 \rangle$ (ζ), deformations of oxygen octahedrons (π) and their counterrotation around the $\langle 111 \rangle$ axis (Ω) [15–17] (Fig. 1a).

Spin states of BiFeO₃ are evaluated from the Hamiltonian

$$H = - \sum_{i<j} J_{ij}(\mathbf{s}_i \cdot \mathbf{s}_j) + \sum_{i<j} \mathbf{D}_{ij}[\mathbf{s}_i \times \mathbf{s}_j] + K_1 \sum_i (\mathbf{s}_i \cdot \mathbf{n}_u)^2 + K_2 \sum_i (\mathbf{s}_i \cdot \mathbf{n}_z)^2 \quad (1)$$

where K_1, K_2 are the constants of orthorhombic magnetic anisotropy, $\mathbf{n}_u, \mathbf{n}_z$ are the unit vectors oriented in the out – of – plane and in – plane directions in a film, $\mathbf{s}_i, \mathbf{s}_j$ are the spins of neighboring iron ions, J_{ij} is the exchange coupling coefficient, $\mathbf{D}_{ij} = -\mathbf{D}_{ji}$ is the Dzyaloshinskii – Moriya (DM) axial vector determined by the Keffer formula [18]

$$\mathbf{D}_{ij} = V_0[\mathbf{R}_{O-i} \times \mathbf{R}_{O-j}] = V_0[\mathbf{d} \times \mathbf{r}_{ij}] \quad (2)$$

where V_0 is the microscopic constant (for BiFeO₃ $V_0 \sim 1.1 \text{ meV}/\text{\AA}^2$ [17]), $\mathbf{R}_{O-i}, \mathbf{R}_{O-j}$ are the vectors connecting the i -th Fe³⁺ ion, in-between O²⁻ ion and the j -th Fe³⁺ ion in the unit cell, \mathbf{r}_{ij} is the radius – vector connecting i -th and j -th Fe³⁺ ions; vector \mathbf{d} determines the displacement of O²⁻ ion from the line connecting Fe³⁺ ions (Fig. 1b). We express vectors $\mathbf{r}_{ij}, \mathbf{d}$ via the crystal distortions (Ω, \mathbf{p}); the unit vectors \mathbf{g}_i oriented along crystallographic directions $\langle 100 \rangle$ and the lattice parameter a [17]:

$$\mathbf{r}_{ij} = a \cdot \mathbf{g}_{\langle 100 \rangle}, \quad \mathbf{d} = -\frac{a}{2}[\Omega \times \mathbf{g}_i] + \mathbf{p} \quad (3)$$

Here the polar vector \mathbf{p} comprises the relative displacements of Fe³⁺ and Bi³⁺ ions (ζ) and the displacements of O²⁻ ions (π) attributed to the deformations of oxygen octahedrons in BiFeO₃, the value of angle of oxygen octahedrons rotation $|\Omega|$ is taken around 14°.

The Hamiltonian (2) corresponds to the free energy

$$F = \frac{1}{2\chi_{\perp}} M_0^2 \mathbf{m}^2 - (\mathbf{m} \cdot [\mathbf{D} \times \mathbf{l}]) + \gamma \mathbf{e}_p \cdot ((\text{grad } \mathbf{l}) \cdot \mathbf{l} - \mathbf{l} \cdot (\text{div } \mathbf{l})) + A(\nabla \mathbf{l})^2 + K_1(\mathbf{n}_u \mathbf{l})^2 + K_2(\mathbf{n}_z \mathbf{l})^2 \quad (4)$$

where $\mathbf{m} = \frac{\mathbf{M}_1 + \mathbf{M}_2}{2M_0}$, $\mathbf{l} = \frac{\mathbf{M}_1 - \mathbf{M}_2}{2M_0}$ are the unit magnetic and antiferromagnetic vectors; $\mathbf{M}_1, \mathbf{M}_2$ are the magnetization vectors of two magnetic sublattices; $M_0 = \frac{1}{2}|\mathbf{M}_1 + \mathbf{M}_2|$ is the saturation magnetization of one of magnetic sublattices; χ_{\perp} is the transversal magnetic susceptibility (for BiFeO₃ $\chi_{\perp} \sim 5 \cdot 10^{-5}$ [5,12]); \mathbf{e}_p is the unit vector oriented in the polar direction along the polarization attributed to the shift of Bi³⁺ and Fe³⁺ ions, $\gamma = \frac{V_0}{a}(\zeta - 2\frac{\pi_{\perp}}{\sqrt{2}})$, ζ is the displacement of Bi³⁺ ion relative to Fe³⁺ ion, π_{\perp} is the projection of vectors π_i on the (111) plane, ($\pi_{\perp} = 0.044 \text{\AA}$ [19], $\gamma = \frac{171}{a^3} = 0.6 \text{ mJ/m}^2$ [20]), A is the constant of the non – uniform exchange interaction (the spin stiffness, for BiFeO₃ $A = 2 \cdot 10^{-12} \text{ J/m}$ [20]); \mathbf{D} is the macroscopic DM vector determined by the formula

$$\mathbf{D} = a^2 V_0 \Omega \quad (5)$$

that can be obtained in the mean field approximation at $\Lambda \gg a$ where Λ is the characteristic length of spatial variation of antiferromagnetic vector $\mathbf{l}(\mathbf{r})$ (see Supplementing Materials), in our case Λ is the length of spin cycloid ($\Lambda = \lambda$).

Minimization of the free energy determined by Eq. (4) on the variable \mathbf{m} using Lagrange method with the additional conditions $\mathbf{m}^2 + \mathbf{l}^2 = 1, (\mathbf{m} \cdot \mathbf{l}) = 0$ leads to

$$F = A(\nabla \mathbf{l})^2 + \frac{\chi_{\perp}}{2} [\mathbf{H}_D \times \mathbf{l}]^2 + \gamma \mathbf{e}_p \cdot ((\text{grad } \mathbf{l}) \cdot \mathbf{l} - \mathbf{l} \cdot (\text{div } \mathbf{l})) + K_1(\mathbf{n}_u \mathbf{l})^2 + K_2(\mathbf{n}_z \mathbf{l})^2 \quad (6)$$

where $\mathbf{H}_D = \frac{\mathbf{D}}{M_0}$ is the Dzyaloshinskii – Moriya (DM) field.

All terms in the expression (6) are of physical sense. The first term stands for the non-uniform exchange energy, the second and the third terms are caused by the energy of the DMI, the latter terms stand for the energy of magnetic anisotropy. The 2nd term $\frac{\chi_{\perp}}{2} [\mathbf{D} \times \mathbf{l}]^2$, the contribution to magnetic anisotropy, arises due to the canting of the magnetic sublattices caused by the DMI; the 3rd term, the Lifshitz invariant, is responsible for inhomogeneous magnetolectric interaction. Due to the presence of the term, linear in $(\text{grad } \mathbf{l})$, the formation of space modulated structure becomes energetically advantageous in comparison with the uniform magnetic state. The inhomogeneous magnetolectric interaction differs from the microscopic DMI ($\mathbf{D}_{ij}[\mathbf{s}_i \times \mathbf{s}_j]$) that acts between the nearest neighbours; Lifshitz invariant depends on the spontaneous electric polarization vector \mathbf{p} (see Supplementing Materials) and can exist only in the crystals without inversion centre [21]. Hereinafter we consider $\mathbf{p} \parallel [111]$, this ferroelectric state is stable at $\frac{\alpha_{12} - \alpha_{11}}{\alpha_{11}} > 0$ where α_{12}, α_{11} are the coefficients of the ferroelectric thermodynamic potential $\Phi_{\text{st}}(\mathbf{P}, \omega, \mathbf{E}) = -|\alpha_1|P^2 + \alpha_{11}P^4 + (\alpha_{12} - \alpha_{11})(P^2 P_{\perp}^2 + \frac{1}{4}P_{\perp}^4)$ [22,23].

Standard cycloid whose plane of spin rotation contains spontaneous polarization has been previously explored in BiFeO₃, hereinafter we proceed to the analysis of novel cycloidal states taking as an example BiFeO₃ film with the developed (110) surface. The geometry of the problem is shown in Fig. 2, we use the reference frame related to the normal to the film ($\mathbf{OX} \parallel [001], \mathbf{OY} \parallel [110], \mathbf{OZ} \parallel [1\bar{1}0]$), $\mathbf{n}_u = [1, 1, 0], \mathbf{n}_z = [1, \bar{1}, 0]$, and determine the position of \mathbf{l} vector by angular variables θ and φ .

To find the stable magnetic configurations we consider the Euler – Lagrange equations

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