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# Ferroelectric polarization in antiferromagnetically coupled ferromagnetic film



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## **ABSTRACT**

We report the influence of interface antiferromagnetic coupling on magnetoelectric properties of ferromagnetic bi-layers. Electric polarization arising at magnetic ingomogeneity in bi-layered ferromagnetic structure with antiferromagnetic coupling at interface in applied magnetic field has been explored. Diagrams representing dependences of electric polarization on magnetic field  $P(H)$  have been constructed for two magnetic field geometries (in-plane and out-of plane fields). It has been found out that P (H) dependences demonstrate non-monotonic behavior. Peculiarities of polarization in an in-plane-oriented magnetic field have been explained by magnetization processes. It has been shown that a variety of magnetic configurations of Bloch, Neel and mixed Bloch–Neel types can be realized in antiferromagnetically coupled film due to cubic anisotropy contribution. In the area of Bloch magnetic configuration electric polarization vanishes. The critical values of magnetic fields suppressing polarization have been estimated.

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#### 1. Introduction

Magnetoelectricity is rapidly developing area of research. Prospects of application of magnetoelectric materials in a wide range of spintronics along with their unconventional physical properties make these compounds attractive for researchers and technologists.

Magnetoelectric effect has been discovered in multiferroic materials whose crystal structure allows coexistence of magnetic and ferroelectric ordering. Later on, the composites representing the combination of magnets and ferroelectrics were considered as perspective magnetoelectric materials. Ferroelectric polarization there basically relies on the ions shift that is expected in the crystals whose symmetry group lacks of space inversion symmetry operation  $[1-3]$  $[1-3]$  $[1-3]$ , magnetic polarization is attributed to transition metal ions.

Nowadays the alternative mechanism of ferroelectric effect related to asymmetric magnetization in magnetoelectrics becomes a topic of hot scientific discussions  $[4-9]$  $[4-9]$  $[4-9]$ . Few years ago, the considerable attention was devoted to spiral-spin multiferroics where electric polarization is magnetically induced due to efficient magnetoelelectric coupling mechanism [\[10](#page--1-0)–[14\]](#page--1-0). Origin of electric

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<http://dx.doi.org/10.1016/j.jmmm.2016.04.058> 0304-8853/© 2016 Elsevier B.V. All rights reserved. polarization was explained appealing to spin-exchange and spinorbit interactions [\[10\]](#page--1-0) and flexomagnetoelectric effect concept [\[11](#page--1-0)-[14,16,17\].](#page--1-0) Recent breakthroughs in magnetoelectricity are related to the discovery of multiferroic properties of cubic magnets (rare earth iron garnets and orthoferrites)  $[4-9]$  $[4-9]$  $[4-9]$ . Lack of the inversion center of spin arrangement along with exchange interactions and spin-orbit coupling between different magnetic ions are considered as the possible mechanisms explaining appearance of electric polarization in these materials.

In the present article we develop the concept of magnetoelectricity appearing in magnetic materials. As a source of ferroelectricity we consider magnetic inhomogeneity at an interface of the layered ferromagnetic structure. It depends on intrinsic magnetic properties of the layers, geometry of a film and external agents. We focus herein on the role of interlayer coupling and explore peculiarities of electric polarization in a case of antiferromagnetic ordering at interface. Layers differ by the type of uniaxial magnetic anisotropy, and due to antiferromagnetic coupling (AFC) nonuniform distribution of magnetization generating electric polarization is established in each of the layers. We consider the overall polarization of the film and explore its behavior in magnetic field dependent on the interlayer exchange and magnetic anisotropy of the layers. We consider two field geometries (magnetic field applied in a film plane (H||[011]) and magnetic field perpendicular to the film  $(H||[100])$ ).

### 2. Model

Magnetization processes and related changes of electric polarization are exemplified on a (100)-oriented film. The geometry of the film is shown in Fig. 1, OX axis is taken to be oriented along normal to a surface, OZ axis is directed along boundary of an interface. Film contains two exchange coupled layers with combined magnetic anisotropy that includes natural cubic and induced uniaxial magnetic anisotropies. The layers differ by the uniaxial component of magnetic anisotropy, the first layer is characterized by induced magnetic anisotropy of the "easy axis" type, the second layer is characterized by induced magnetic anisotropy of the "easy plane" type. Suppose that magnetic field is applied along "easy" magnetic directions and consider two situation of in-plane (H|| [011]) and out-of-plane (H||[100]) oriented magnetic fields. For a sake of convenience, we transform coordinate frame rotating ZOY plane in such a way that OY axis becomes parallel to the [011] "easy" direction in (100) plane.

The density of free energy is taken in the form

$$
E = \sum_{i=1}^{2} \left[ A \left[ \left( \frac{\partial M_{ix}}{\partial x} \right)^{2} + \left( \frac{\partial M_{iy}}{\partial y} \right)^{2} + \left( \frac{\partial M_{iz}}{\partial z} \right)^{2} \right] + K_{c} \left( M_{ix}^{2} M_{iy}^{2} + M_{ix}^{2} M_{iz}^{2} + M_{iy}^{2} M_{iz}^{2} \right) + K_{i} \left( \mathbf{M}_{i} \mathbf{n} \right)^{2}
$$

$$
- \mathbf{M}_{i} \mathbf{H} + J \mathbf{M}_{1} \mathbf{M}_{2} + \gamma \tilde{\mathbf{P}}_{i} \left( \left( \mathbf{M}_{i} \partial_{x} \mathbf{M}_{i} \right) - \mathbf{M}_{i} \left( \partial_{x} \mathbf{M}_{i} \right) \right) + \frac{\tilde{p}_{ix}^{2} + \tilde{p}_{iy}^{2} + \tilde{p}_{iz}^{2}}{2 \chi_{e}} - \tilde{\mathbf{P}}_{i} \mathbf{E} \right]
$$

where A is the constant of non-uniform exchange interaction,  $K_1$  is the constant of magnetic anisotropy of the "easy axis" type,  $K_2$  is the constant of magnetic anisotropy of the "easy plane" type,  $K_c$  is the constant of cubic magnetic anisotropy ( $K_c < 0$ ), J is the constant of interlayer exchange interaction,  $\tilde{P}$  is the polarization vector, *<sup>χ</sup><sup>e</sup>* is the dielectric susceptibility, *<sup>γ</sup>* is the coefficient of nonuniform magnetoelectric interaction,  $H$  is the applied magnetic field,  $E$  is the electric field.

Equilibrium values of local magnetization and electric polarization were determined numerically by minimization of the free energy density functional [\[15\].](#page--1-0) To conduct simulation we used the parameters of iron garnets which can vary in sufficiently wide range, we take  $A=10^{-7}$  erg/cm,  $M_{01}=36$  G,  $M_{02}=70$  G,  $|K_1|,|K_2|,|K_3|$  $K_c$ |∼(10<sup>3</sup>–10<sup>5</sup>) erg/cm<sup>3</sup>,  $\gamma$  = 10<sup>-6</sup> erg/cm<sup>2</sup>.

It should be noted that in the case when cubic magnetic anisotropy is taken into account the vector of electric polarization has two components  $\tilde{P}_y$ ,  $\tilde{P}_z$  [\[16\].](#page--1-0) To obtain the magnitude of overall polarization of a film we make a summation of the local polarizations and calculate the length of polarization vector  $\tilde{P} = \sqrt{\tilde{P}_y^2 + \tilde{P}_z^2}$ . Below we discuss the results of simulation for two orientations of magnetic field H|| 100], H||011].

#### 3. Results and discussion

The dependences of overall polarization  $P = \frac{\tilde{P}}{x e^{\gamma}}$  on magnetic







Fig. 2. Dependences of electric polarization on magnetic field at different values of interlayer coupling parameters (a)  $H||[100]$ , (b)  $H||[011]$ , blue curve 1 stands for  $J=1$ , red curve 2 stands for J=0.6, green curve 3 stands for J=0.1,  $K_1$ =2·10<sup>4</sup> erg/cm<sup>3</sup>  $K_2 = -7.10^4$  erg/cm<sup>3</sup>,  $K_c = -2.10^4$  erg/cm<sup>3</sup>. (For interpretation of the references to color in this figure legend,the reader is referred to the web version of this article.)

field for the cases  $H||[100]$  and  $H||[011]$  are shown in Fig. 2a, b. It is seen that electric polarization that exists in AFC bi-layers is supported by applied magnetic field. Polarization increases with increase of magnetic field, after attaining the maximum value polarization shrinks with further enhancement of magnetic field and drops to zero at the critical field ( $H_{cr}$ ). The value of critical field  $H_{cr}$ can be regulated by the strength of interlayer exchange coupling. Fig. 2 shows that in the case when interlayer antiferromagnetic coupling is strong enough  $(I=1)$  electric polarization exists in a wide range of magnetic fields. To supress polarization high magnetic fields (of the orders of 60 kOe) are required.

It is seen in Fig. 2b that deflection or "cavity" on  $P(H)$  curve appears in magnetic field  $H$ ||[011] at  $|K_2|>|K_1|$ . The origin of the "cavity" is attributed to rotation of magnetization vector which sticks out from the rotational plane  $(X'OY)(0\bar{1}1)$  (see Fig. 1) in a vicinity of interface. Plots showing dependences of angles determining the position of vector  $\mathbf{m}$  ( $\mathbf{m} = \frac{M}{M_s} = (\sin \theta \cos \varphi, \sin \theta \sin \varphi, \cos \theta)$ ) in XYZ coordinate frame on magnetic field H||[011] are presented in [Fig. 3.](#page--1-0) Magnetic field induces a variety of magnetic configurations in AFC bi-layer: a Neel-like magnetic distribution (the linear regions on  $\theta$ (H)) plots at  $\varphi \neq 90^0$ ), the combination of Bloch and Neel walls, and as we show below Bloch walls. It is seen in [Fig. 3a](#page--1-0) that the polar Fig. 1. Geometry of a problem.  $\Box$  angle  $\theta$  deviates from  $90^{\circ}$  (magnetization sticks out from the

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