ISSN 0031-918X, Physics of Metals and Metallography, 2020, Vol. 121, No. 6, pp. 526–531. © Pleiades Publishing, Ltd., 2020. Russian Text © The Author(s), 2020, published in Fizika Metallov i Metallovedenie, 2020, Vol. 121, No. 6, pp. 583–588.

# ELECTRICAL AND MAGNETIC = PROPERTIES

# Hysteresis of the Electric Polarization in a Two-Layer Ferromagnetic Film with a Vortical Distribution of Magnetization

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Received December 17, 2019; revised January 28, 2020; accepted January 31, 2020

**Abstract**—Hysteresis of the electric polarization of a two-layer exchange-coupled ferromagnetic film whose layers have the easy-plane and easy-axis anisotropy upon magnetization reversal is studied numerically. The magnetization reversal was carried out by a magnetic field perpendicular to the film plane from the saturation state along the easy magnetization axis. The dependences of the average electric polarization and the reduced magnetization of the film layers on the external magnetic field strength are constructed. The feasibility of sign reversal of the electric polarization in a magnetic field in films whose thickness is smaller than the transverse dimensions is found. No polarization sign reversal is observed in thicker films upon magnetization reversal.

*Keywords:* two-layer ferromagnetic film, inhomogeneous magnetoelectric effect, electric polarization, hysteresis

DOI: 10.1134/S0031918X20060137

#### **INTRODUCTION**

The study of the conditions for the rise of electric polarization on a vertical magnetic structure is of scientific and practical interest. Magnetic irregularities in the form of static vortices, forming under various conditions in ferromagnetic nanosized thin films, can be used in the rapidly developing field of high-density information carriers [1] and in spintronics devices [2]. A magnetic vortex with a nanometer-sized core is described by two topological quantities [3, 4]. One of them is chirality, i.e., the direction of rotation of the magnetic moment in the plane: clockwise or anticlockwise. The other quantity is the polarity, which is determined by the direction in which the magnetization emerges from the vortex plane: up or down.

An important task is to study the conditions for the rise of a vortex structure, as well as the possibilities of controlling its states [5]. Vortex nucleation and stabilization can occur in a ferromagnetic film under the action of an external magnetic field [6], on a magnetic irregularity [7, 8], and in a multilayer film [9]. The properties of the vortex also depend on the shape and size of the sample on which it is formed and the history of its magnetization [10].

The switching between the described vortex states can be carried out in various ways, particularly when the inhomogeneous magnetoelectric effect is manifested [11] or by an electric field [12]. The study of the features of electric polarization on magnetic irregularities in ferrite garnet films [13–15] is of certain interest. The conditions for the rise of polarization on a magnetic irregularity near the interlayer boundary of a similar ferrite garnet film were studied in [16] under the assumption that its transverse dimensions are much larger than the thickness and the magnetization in the film plane is uniformly distributed.

In this article, we study numerically the features of the electric polarization of a film, arising on a vertical irregularity of a two-layer ferrite garnet film upon a magnetization reversal by an external magnetic field perpendicular to the film plane. Hysteresis of electric polarization and magnetization in films of various thickness is calculated in detail.

### 1. STATEMENT OF THE PROBLEM

We consider a two-layer film whose upper layer has an easy-axis anisotropy and whose lower layer has an easy-plane anisotropy. The film has finite sizes and a square cross section; its dimensions vary from 150 to 550 nm and thickness varies from 100 to 350 nm (see Figs. 2 and 5). The z axis coincides with the axis of uniaxial anisotropy. The external magnetic field is parallel to the z axis. The energy functional of the system is

$$W = \sum_{i=1}^{2} \int_{V_{i}} dV \{ E_{u,i} + E_{H,i} + E_{d,i} + E_{ex,i} \} - \int_{S} E_{int} dS.$$
(1)

It includes the energy of uniaxial magnetic anisotropy of the sample  $E_{u,i} = \frac{K_i}{M_i^2} \mathbf{M}_{z,i}^2$ , the Zeeman energy  $E_{\mathrm{H},i} = -\mathbf{M}_i \mathbf{H}$ , the dipole interaction energy  $E_{\mathrm{H},i} = -\mathbf{M}_i \mathbf{H}$ , the exchange interaction energy  $E_{\mathrm{ex},i} = \frac{\alpha_i}{2M_i^2} \times \mathbf{I}_{\mathrm{ex},i}$ 

 $\left[\left(\frac{\partial \mathbf{M}_i}{\partial x}\right)^2 + \left(\frac{\partial \mathbf{M}_i}{\partial y}\right)^2 + \left(\frac{\partial \mathbf{M}_i}{\partial z}\right)^2\right], \text{ and the interlayer}$ exchange interaction energy  $E_{\text{int}} = \frac{J}{M_1 M_2} \mathbf{M}_1 \mathbf{M}_2.$ Here,  $K_i$  are the uniavial axis to the universe of the unive

Here,  $K_i$  are the uniaxial anisotropy constants of the layers,  $M_i$  are the saturation magnetizations of the layers, **H** is the external magnetic field,  $\mathbf{H}^{(m)}$  is the magnetic dipole interaction field,  $\alpha_i$  are the exchange interaction constants, and J is the interlayer exchange interaction constant.

# 2. METHOD OF CALCULATION

The problem of finding the equilibrium state was solved numerically. The calculations were performed using the OOMMF 3D modeling software package [17] with the discretization on a rectangular grid with a step of 5 nm in the *x* and *y* coordinates and 3 nm in the *z* coordinate. The calculation was performed for a two-layer film whose parameters are typical of a ferrite garnet film:  $M_1 \approx 30$  G,  $M_2 \approx 70$  G,  $\alpha \approx 10^{-7}$  erg/cm,  $K_1 \approx 2 \times 10^4$  erg/cm<sup>3</sup>,  $K_2 \approx -7 \times 10^4$  erg/cm<sup>3</sup>, and J = 1 cm<sup>-1</sup>. For a nonuniform magnetization distribution, the electric polarization vector **P** was calculated by the formula [18]

$$\mathbf{P} = \gamma \chi_{e} [(\mathbf{M} \nabla) \mathbf{M} - \mathbf{M} (\nabla \mathbf{M})], \qquad (2)$$

where  $\chi_e$  is the electric polarizability and  $\gamma$  is the coefficient of inhomogeneous magnetoelectric interaction. Next, we calculate the polarization vector related to the product of these quantities and the square of the saturation magnetization:  $\mathbf{P} \rightarrow \mathbf{P}/\gamma \chi_e M_i^2$ ; the dimension of the reduced polarization is cm<sup>-1</sup>. The average polarization is calculated by the formula:

$$\overline{\mathbf{P}} = \frac{1}{V} \int_{V} \mathbf{P}(x, y, z) \, dx \, dy \, dz. \tag{3}$$

# 3. DISCUSSION OF THE RESULTS

Let us consider the change in the average polarization upon magnetization reversal of a two-layer film



**Fig. 1.** (a) Mean polarization vs. the magnitude of the external magnetic field. (b) Reduced magnetization of the layers vs. the magnitude of the external magnetic field. Sample of  $200 \times 200 \times 120$  nm.

from the saturation state along the z axis to the saturation state against the z axis (curves  $\alpha$  in Figs. 1 and 4) and then from the saturation state against the z axis to the saturation state along the z axis (curves  $\beta$  in Figs. 1) and 4). Significant distinctions were found in the dependences of the average polarization on the field for films whose thickness is smaller (Fig. 1a) and greater (Fig. 4a) than the transverse dimensions. Let us consider the first case. It can be seen that, except for a small range of fields near zero, the average polarization of the film remains positive. It is contributed mainly by the polarization of the layer with the easyplane anisotropy; the average polarization of the layer with the easy-axis anisotropy is negative and smaller in magnitude. Figure 1b shows the corresponding dependences of the reduced magnetization: curves  $\alpha'$  and  $\beta'$ correspond to the easy-axis layer; the hysteresis loop here is rectangular. Curves  $\alpha$ " and  $\beta$ " correspond to the easy-plane layer; the hysteresis loop here reaches the saturation magnetization in large fields. As the saturating field decreases, the average polarization

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**Fig. 2.** Magnetization distribution at the upper and lower boundaries of the film. External magnetic field H = (a) 1760, (b) 503, (c) -276, and (d) -1760 Oe. Sample of 200 × 200 × 120 nm.

increases. In this case, the magnetization deviates into sample (Fig. 2a). After reaching a local maximum of the polarization curve at point *I* in Fig. 1a (this corresponds to the magnetization distribution at the upper and lower boundaries of the film in Fig. 2a), in the layer with easy-plane anisotropy, a vortical magnetization distribution begins to form (Fig. 2b). In Fig. 1b, this corresponds to the beginning of the divergence of curves  $\alpha'$  and  $\alpha''$ . In the region from the saturation field to point 2 (Fig. 1a), the dependences of the average polarization  $\alpha$  and  $\beta$  are superimposed on each other. At point 2, the vortical magnetization distribution also begins to form in the layer with easy-axis anisotropy.

At point 3, in a field of about -250 Oe,  $\overline{P}_z$  is negative. This corresponds to the state in which the easyplane magnetization of the major part of the anisotropic layer is already oriented along the field but, in the vortex core, there is still magnetization oriented mainly along the z axis (Fig.2c), which leads to a sharp decrease in the average polarization. The corresponding polarization distributions near the lower and upper boundaries of the film are shown in Figs. 3a and 3b. It can be seen that, near the lower boundary of the film, the maximum  $P_z$  at the center of the film is greater than zero, but its absolute value is about half the minimum  $P_z$  near the lower boundary. At the periphery of the film,  $P_z$  is smaller than zero. As a result, the average polarization of the upper layer of the film will be greater in the absolute value than the mean polarization of the lower layer. Hence, since the average polarization of the upper layer of the film is negative, the average polarization of the film becomes negative. Thus, by varying the magnetic field, it is possible to change the sign of the electric polarization of the film. The magnetization reversal process is strongly affected by the layer with the easy-axis anisotropy, in which, as can be seen in dependences  $\alpha'$  and  $\alpha''$  in Fig. 2, the magnetization is reversed first.

Let us now consider (see Figs. 3c and 3d) what happens to the polarization distribution near the upper and lower boundaries of the film after the magnetization in the vortex core in both layers is oriented along the field (point 4 in Fig. 1). In this case, the

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**Fig. 3.** Polarization component  $P_z$  vs. x and y coordinates. External magnetic field H = -250 Oe (a) near the lower boundary and (b) near the upper boundary of the film. External magnetic field H = -400 Oe (c) near the lower boundary and (d) near the upper boundary of the film. Sample of  $200 \times 200 \times 120$  nm.

polarization is greater than zero almost everywhere near the lower boundary and its absolute value is almost an order of magnitude greater than the minimum polarization near the lower boundary of the film; as a result, the average polarization of the film becomes positive and increases sharply. At point 5 in Fig. 1, the second maximum of average polarization is found—this time, in a negative field. In this case, the magnetization is oriented outward (Fig. 2d).

The same regularities in the behavior of the average polarization and magnetization are observed upon the reverse magnetization reversal (curve  $\beta$ ). The average polarization reaches its minimum (point *6*) in a positive field of about 250 Oe.

The smaller the sample sizes, the smaller the magnitude of the field in which of the polarization reaches local maxima, since the magnetization rotates faster. In this case,  $\overline{P}_z$  is greater. Correspondingly, the smaller the sample sizes, the smaller the minimum of the average polarization near the zero field.

Let us now consider the specific features of the dependence of the average polarization on the field for films whose thickness exceeds the transverse sizes. In particular, for a  $200 \times 200 \times 300$ -nm sample, near the zero field, an increase in the average polarization is first observed, which is then replaced with a decrease (see Fig. 4a). Figure 5 shows the corresponding magnetization distributions upon the magnetization reversal of the film from the saturation state against the z axis to the saturation state along the z axis (see curves  $\beta$  in Fig. 4). In the absence of an external field, when the average polarization reaches its minimum at point 1, the vortex distribution of magnetization can form only in a layer with the easy plane anisotropy, while, in a layer with the easy axis anisotropy, the magnetization is oriented against the z axis (Fig. 5a). Then, in a positive field, a growth is observed (see point 2 in Fig. 4a). This is due to the formation of a vortex in a layer with the easy-axis anisotropy (Fig. 5b). With a further increase in the magnetic field,  $\overline{P}_z$  decreases (see point 3



**Fig. 4.** (a) Mean polarization vs. the strength of the external magnetic field. (b) Reduced magnetization of the layers vs. the strength of the external magnetic field. Sample of  $200 \times 200 \times 300$  nm.

in Fig. 4), which is ascribed to the reorientation of the magnetization toward field, described above (Fig. 5c).

#### CONCLUSIONS

A numerical study of the change in electric polarization upon magnetization reversal of a two-layer exchange-coupled ferromagnetic film, whose layers have an easy-plane and easy-axis anisotropy, by an external magnetic field perpendicular to the film plane, has been completed. The magnetization reversal was carried out from the saturation state along the z axis (perpendicular to the film plane and coinciding with the easy axis) and vice versa.

Significant difference was found in the behavior of the dependences of the average electric polarization for films of various thicknesses. For samples whose thickness is smaller than the transverse sizes, hysteresis of the electric polarization with the polarization sign reversal is observed. The following regularities have been found. With a decrease in the saturating field, the average polarization increases. In this case,



**Fig. 5.** Magnetization distribution at the upper and lower boundaries of the film. External magnetic field H = (a) 0, (b) 603, and (c) 628 Oe. Sample of  $200 \times 200 \times 300$  nm.

the magnetization deviates inward in a positive magnetic field or outward in a negative field. In magnetic fields, after reaching a local maximum of the polarization curve in a layer with the easy-plane anisotropy, a vortical magnetization distribution begins to form. With a further decrease in the magnetic field, a vortical magnetization distribution also begins to form in the layer with the easy-axis anisotropy. The minimum in the dependence of the average polarization upon magnetization reversal from the saturation state along the easy magnetization axis is found in a negative field. The minimum corresponds to such a distribution of magnetization in which, in a major part of the layer with the easy-plane anisotropy, the magnetization is already oriented along the field but, in the vortex core, the magnetization oriented in the opposite direction is still present. Similar features are also observed upon the opposite magnetization reversal.

A different behavior of the average polarization upon magnetization reversal is observed in films whose thickness is comparable to or greater than the transverse sizes of the layers. In this case, hysteresis of the electric polarization is pronounced weakly. Near the zero field, only a slight increase in the average polarization is observed, which is then replaced with a decrease. In the absence of an external field, when the mean polarization reaches its minimum, a vortical magnetization distribution can form only in a layer with easy-plane anisotropy, while, in a layer with easyaxis anisotropy, the magnetization is oriented almost against the z axis. Then, with an increase in the absolute value of the magnetic field, an increase of  $\overline{P}_{z}$  is observed. This is explained by the formation of a vortex structure in a layer with easy-axis anisotropy. With a further increase in the magnetic field,  $P_{z}$  decreases due to the orientation of the magnetization along the field.

#### REFERENCES

- D. G. Geng and Y. M. Jin, "Magnetic vortex racetrack memory," J. Magn. Magn. Mater. 423, 84–89 (2017).
- 2. G. A. Prinz, "Magnetoelectronics," Science 282, 1660–1663 (1998).
- T. Shinjo, T. Okuno, R. Hassdorf, K. Shigeto, and T. Ono, "Magnetic vortex core observation in circular dots of permalloy," Science 289, 930–932 (2000).
- R. Moriya, L. Thomas, M. Hayashi, Y. B. Bazaliy, Ch. Rettner, and S. P. Parkin, "Probing vortex-core dynamics using current-induced resonant excitation of a trapped domain wall," Nat. Phys. 4, 368–372 (2008).
- P. I. Karpov and S. I. Mukhin, "Polarizability of electrically induced magnetic vortex plasma," Phys. Rev. B 95, 195136-1–195136-16 (2017).
- J. Li, Y. Wang, J. Cao, X. Meng, F. Zhu, and R. Tai, "The control of magnetic vortex state in rectangular nanomagnet," J. Magn. Magn. Mater. 451, 379–384 (2018).

- G. A. Meshkov, A. P. Pyatakov, A. D. Belanovsky, K. A. Zvezdin, A. S. Logginov, "Writing vortex memory bits using electric field," J. Magn. Soc. Jpn. 36, 46– 48 (2012).
- N. V. Shul'ga and R. A. Doroshenko, "Nonuniform magnetoelectric effect in a nano-sized ferromagnetic film with surface anisotropy," Phys. Met. Metallogr. 120, 639–645 (2019).
- N. V. Shul'ga and R. A. Doroshenko, "Electric polarization in two-layer bounded ferromagnetic film," J. Magn. Magn. Mater. 471, 304–309 (2019).
- Z.-H. Wei, Ch.-R. Chang, N. A. Usov, M.-F. Lai, and J. C. Wu, "Evolution of vortex states under external magnetic field," J. Magn. Magn. Mater. 239, 1–4 (2002).
- V. G. Bar'yakhtar, V. A. L'vov, and D. A. Yablonskii, "Theory of non-uniform magnetoelectric effect," Pis'ma Zh. Eksp. Teor. Fiz. 37, 565–567 (1983).
- A. P. Pyatakov, A. S. Sergeev, E. P. Nikolaeva, T. B. Kosykh, A. V. Nikolaev, K. A. Zvezdin, and A. K. Zvezdin, "Micromagnetism and topological defects in magnetoelectric media," Phys.-Usp. 58, 981– 992 (2015).
- I. S. Veshchunov, S. V. Mironov, W. Magrini, V. S. Stolyarov, A. N. Rossolenko, V. A. Skidanov, J.-B. Trebbia, A. I. Buzdin, Ph. Tamarat, and B. Lounis, "Direct evidence of flexomagnetoelectric effect revealed by single-molecule spectroscopy," Phys. Rev. Lett. 115, 027601 (2015).
- 14. G. V. Arzamastseva, A. M. Balbashov, F. V. Lisovskii, E. G. Mansvetova, A. G. Temiryazev, and M. P. Temiryazeva, "Properties of epitaxial (210) iron garnet films exhibiting the magnetoelectric effect," J. Exp. Theor. Phys. **120**, 687–701 (2015).
- D. P. Kulikova, T. T. Gareev, E. P. Nikolaeva, T. B. Kosykh, A. V. Nikolaev, Z. A. Pyatakova, A. K. Zvezdin, and A. P. Pyatakov, "The mechanisms of electric field-induced magnetic bubble domain blowing," Phys. Status Solidi RRL 12, 1800066 (2018).
- Z. V. Gareeva, R. A. Doroshenko, N. V. Shulga, and K. Harbusch, "Peculiarities of electric polarization in bi-layered longitudinally magnetized ferromagnetic film," J. Magn. Magn. Mater. **321**, 1163–1166 (2009).
- M. J. Donahue and D. G. Porter, *OOMMF User's Guide. Version 1.0 NISTIR 6376* (National Institute of Standards and Technology, Gaithersburg, 1999).
- M. Mostovoy, "Ferroelectricity in spiral magnets," Phys. Rev. Lett. 96, 067601 (2006).

Translated by E. Chernokozhin