Physics Letters A 382 (2018) 1124–1130

Contents lists available at ScienceDirect

Physics Letters A

www.elsevier.com/locate/pla

Discussion

The converse magnetoelectric coupling in asymmetric granule/matrix composite film with Ni/PZT component

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ARTICLE INFO

Article history:
Received 19 December 2017
Received in revised form 1 February 2018
Accepted 25 February 2018
Available online 6 March 2018
Communicated by M. Wu

Keywords:
Magnetoelectricity
Electric polarization
Magnetization
Magnetic anisotropy

ABSTRACT

In this work, a type of asymmetric granule/matrix composite film is designed, where the Ni granule is dispersed in PZT matrix, meanwhile the top and bottom electrode is constituted by Au and SRO respectively. Predicted through the electrostatic screening model and mean field approximation, considerable electrostatic charge is induced on Ni granule surface by ferroelectric PZT polarization. Predicted through the spin splitting model and spherical shell approximation, both the magnetization and magnetic anisotropy of Ni granule are modulated by ferroelectric PZT polarization. As the volume fraction of Ni granule is increased, the electric modulation of magnetization and magnetic anisotropy is reduced and enhanced respectively. As the dimension of granule/matrix composite is varied, such modulation is retained. Due to the large area–volume ratio of nano-granule, this work benefits to realize the converse magnetoelectric coupling in nanoscale.

1. Introduction

In the magnetic/dielectric composite film, the magnetism could be manipulated by electric polarization, which is known as the converse magnetoelectric (CME) coupling. Usually, there are two types of CME coupling, i.e. the strain-mediated and charge-mediated type. During the strain-mediated coupling, the electric polarization induces electrostrictive strain in dielectric component, and the piezomagnetism in magnetic component is generated through strain transfer [1–3]. During the charge-mediated coupling, the electric polarization induces electrostatic charge on the magnetic/dielectric interface, and the itinerant magnetism on interface is influenced by the spin-polarizing of electrostatic charge [4–6]. Based on the CME coupling, the electric-write magnetic-read memory device could be realized, which increases the running speed and decreases the energy loss in computing machine [7]. Thus, the CME coupling is attractive in the condensed matter physics and functional material.

At present, the CME coupling is mainly investigated in laminate composite film, where the magnetic and dielectric laminates are combined layer by layer. In such composite structure, the strength of strain transfer, and the depth of screening region, is crucial to the strain-based and charge-based CME coupling respectively. On the magnetic/dielectric interface, the strain transfer is seriously restricted by substrate, while the screening region is only as deep as 0.1∼1.0 nm [8,9]. Consequently, the CME coupling is usually weakened.

Compared to the laminate composite film, stronger CME coupling could be obtained in the granule/matrix composite film. Such composite film is assembled by distributing magnetic granule into dielectric matrix, where the granule size is in nanoscale [10–12]. Due to the large area–volume ratio of magnetic granule, the CME coupling on the granule/matrix interface is obvious. Furthermore, the size effect in magnetic granule is significant. If the granule size is larger or smaller than the critical size of superparamagnetism, the magnetic granule exhibits ferromagnetism and superparamagnetism respectively [13]. Consequently, novel CME coupling may exist in granule/matrix composite film.

However, to achieve meaningful CME coupling in granule/matrix composite film, the composite structure is critical. Usually, the granule/matrix composite is symmetric type, i.e. mirror symmetry with the middle horizon plane. As the electric polarization orients upward and downward, its manipulation of magnetism in granule/matrix composite is identical. In order to the bipolar CME coupling that does not cancel out, i.e. the distinctly different manipulation of magnetism between upward and downward ferroelectric polarization, the asymmetric granule/matrix composite is required.

In this work, a type of asymmetric granule/matrix composite film is designed, where the screening depth of top and bot-

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https://doi.org/10.1016/j.physleta.2018.02.032
0375-9601/

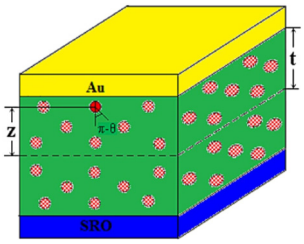

Fig. 1. (Color online.) The illustration to granule/matrix composite structure with Ni/PZT component.

tom electrodes is distinctly different from each other. The component of top and bottom electrode is Au and $SrRuO_3$ (SRO) respectively. The dielectric matrix is constituted by the ferroelectric oxide $PbZr_xTi_{1-x}O_3$ (PZT). The magnetic granule is formed by the ferromagnetic metal Ni, meanwhile the granule shape is defined as regular sphere. Through theoretical model, the CME coupling in present granule/matrix composite film is investigated. Under the ferroelectric polarization of PZT matrix, both the magnetization and magnetic anisotropy of Ni granule is modulated. Furthermore, the CME coupling in present granule/matrix composite film is bipolar type.

2. Theoretical model

2.1. Electrostatic screening

The composite structure is illustrated in Fig. 1. Usually the SRO substrate is adopted as bottom electrode. Both the SRO and PZT crystalline is perovskite type, which permits the preferred orientation of (001) PZT. Thus the in-plane epitaxial strain of PZT matrix is estimated by $u = [a_{PZT} - a_{SRO}]/a_{PZT} = -2.2\%$, where the lattice constants are $a_{PZT} = 0.402$ nm and $a_{SRO} = 0.393$ nm [14]. Under such compressive strain, the spontaneous polarization of PZT matrix is along the out-of-plane axis [15]. Accordingly, the upward and downward polarization is defined as $P_0 < 0$ and $P_0 > 0$ respectively. Based on Thomas–Fermi model, the potential drop across out-of-plane polarized PZT matrix is expressed as [16,17]:

$$\Psi_{\infty}^{(0)} = \frac{(\delta_{SRO} - \delta_{Au})t - (\delta_{SRO} + \delta_{Au})z}{\varepsilon_r(\delta_{SRO} + \delta_{Au}) + 2t}\frac{P_0}{\varepsilon_0} \tag{1}$$

where t represents the half thickness of PZT matrix, the screening depths are $\delta_{Au} = 0.06$ nm and $\delta_{SRO} = 0.6$ nm [16], the spontaneous polarization and static dielectric constant is $P_0 \approx \pm 40$ μC/cm^2 and $\varepsilon_r \approx 100$ respectively [18].

If one Ni granule is solely embedded in PZT matrix, the electric polarization of PZT matrix is expressed by $\vec{P} = \vec{P}_0 + \varepsilon_r\varepsilon_0[-\vec{\nabla}\Psi^{(0)}]$, where $\Psi^{(0)}$ represents the electrostatic screening potential [16]. The spontaneous polarization $\vec{P}_0$ is uniaxially along the out-of-plane axis. The other item $\varepsilon_r\varepsilon_0[-\vec{\nabla}\Psi^{(0)}]$ represents the extra polarization induced by electrostatic screening field $-\vec{\nabla}\Psi^{(0)}$. Near Ni/PZT interface, the local electrostatic screening field is non uniaxial. Consequently, the polarization $\vec{P}$ near Ni/PZT interface deviates from the uniaxial polarization $\vec{P}_0$. Under $P_0 < 0$ and $P_0 > 0$, the average potential in PZT matrix is negative and positive respectively, as demonstrated in Eq. (1). Such potential induces the accumulated or depleted carrier on Ni granule surface, which is demonstrated through the surface charge density $\sigma_{\delta}^{(0)}$.

A spherical coordinate is set up around the Ni granule, where the polar axis is along the vertical direction. Outside the spherical surface, the Laplace equation $\nabla^2\Psi^{(0)} = 0$ is adopted for insulating PZT matrix. Furthermore, as the radial position r becomes infinite, the electrostatic potential $\Psi^{(0)}$ tends to $\Psi_{\infty}^{(0)}$ expressed in Eq. (1). Thus, the electrostatic potential $\Psi^{(0)}$ is deduced as:

$$\Psi^{(0)} = \Psi_{\infty}^{(0)} + \frac{\partial\Psi_{\infty}^{(0)}}{\partial z}r\cos\theta + \sum_{n=0}^{\infty}\frac{R^{n+1}}{r^{n+1}}\Psi_n^{(0)}P_n(\cos\theta) \tag{2}$$

where R represents the spherical radius of Ni granule, $P_n(\cos\theta)$ is the n-order Legendre polynome, and $\Psi_n^{(0)}$ is constant potential. Inside the spherical surface, the electrostatic potential $\Psi_{\delta}^{(0)}$ is expressed by the Thomas–Fermi model [16,17]:

$$\nabla^2\Psi_{\delta}^{(0)} = -\frac{1}{\varepsilon_0}\frac{\sigma_{\delta}^{(0)}}{\delta_{Ni}}\exp\left(\frac{r-R}{\delta_{Ni}}\right) \tag{3}$$

where δ_{Ni} represents the screening depth of Ni granule. Applying Gauss law and potential continuity on Ni/PZT interface, the charge density $\sigma_{\delta}^{(0)}$ is derived as:

$$\begin{aligned}\sigma_{\delta}^{(0)} = {} & \frac{\varepsilon_r[(\delta_{Au} - \delta_{SRO})t + (\delta_{Au} + \delta_{SRO})z]}{[R + \varepsilon_r\delta_{Ni}][\varepsilon_r(\delta_{Au} + \delta_{SRO}) + 2t]}P_0 \\ & + \frac{2[\varepsilon_r(\delta_{Au} + \delta_{SRO}) - t]R}{[R + 2\varepsilon_r\delta_{Ni}][\varepsilon_r(\delta_{Au} + \delta_{SRO}) + 2t]}P_0\cos\theta\end{aligned} \tag{4}$$

where $\sigma_{\delta}^{(0)}$ varies with both z and $\cos\theta$ linearly.

Actually, numerous Ni granules are embedded in PZT matrix, as illustrated in Fig. 1. Between adjacent Ni granule surfaces, the electrostatic screening interacts with each other, which is described through the perturbation method and mean field approximation. Under the zero-level condition, i.e. the isolated condition, the charge density $\sigma_{\delta}^{(0)}$ on each Ni granule surface has been expressed in Eq. (4). It is equivalent to some free charge in PZT matrix, which is expressed as:

$$\rho_e = \eta\frac{\oiint\sigma_{\delta}^{(0)}R^2\sin\theta d\theta d\varphi}{4\pi R^3/3} \tag{5}$$

where ρ_e is the free charge density, and η represents the volume fraction of Ni granule in composite. Correspondingly, the potential drop in PZT matrix obeys the Poisson equation $\nabla^2\Psi_{\infty} = -\rho_e/(\varepsilon_r\varepsilon_0)$, which is deduced as:

$$\Psi_{\infty} = \Psi_{\infty}^{(0)} + \eta\left[\Psi_{\infty,0} + \Psi_{\infty,1}\left(\frac{z}{t}\right) + \Psi_{\infty,2}\left(\frac{z}{t}\right)^2 + \Psi_{\infty,3}\left(\frac{z}{t}\right)^3\right] \tag{6}$$

Here, the $\Psi_{\infty,0}$, $\Psi_{\infty,1}$, $\Psi_{\infty,2}$ and $\Psi_{\infty,3}$ are constant potential drops, which represent the first-level modification for $\Psi_{\infty}^{(0)}$.

Outside the spherical surface, the electrostatic potential Ψ still tend to Ψ_{∞}, as the radial position r becomes infinite. Thus, the first-level modification for $\Psi^{(0)}$ is deduced as:

$$\Psi = \Psi^{(0)} + \eta\sum_{i=0}^{3}\Psi_{\infty,i}\left(\frac{z + r\cos\theta}{t}\right)^i + \eta\sum_{n=0}^{\infty}\frac{R^{n+1}}{r^{n+1}}\Psi_n P_n(\cos\theta) \tag{7}$$

where Ψ_n is constant potential drop. Based on Eq. (3) and Eq. (7), applying the Gauss law and potential continuity on Ni granule surface, the first-level modification for $\sigma_{\delta}^{(0)}$ is derived as:

$$\begin{aligned}\sigma_{\delta} = {} & \sigma_{\delta}^{(0)} + \eta\sum_{n=0}^{3}n\sigma_n\left(\frac{z + R\cos\theta}{t}\right)^{n-1}\cos\theta \\ & + \eta\sum_{n=0}^{3}(n+1)\sigma_{z,n}P_n(\cos\theta)\end{aligned} \tag{8}$$

where σ_n is constant surface charge density, while $\sigma_{z,n}$ is surface charge density varying with z. The first and other item of σ_{δ} represents the harmonic and anharmonic surface charge density respectively, which varies linearly and nonlinearly with $\cos\theta$ respectively.