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Electric-field control of exchange bias field in a $Mn_{50.1}Ni_{39.3}Sn_{10.6}/$ piezoelectric laminate



ALLOYS AND COMPOUNDS

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1. Introduction

Electric-field control of magnetization, which is defined as the converse magnetoelectric (CME) effect, has many potential applications in magnetic memory storage, sensors and spintronics [1-3]. Up to now, many studies about CME effect are focused on the piezoelectric/magnet bilayer structures, in which the generated strain from electric field can transfer to the magnetic layer, and thus, affect the magnetism. This strain-mediated effect has been actively studied in many systems, demonstrating the manipulation of magnetic properties such as coercivity (H_c) , domain wall propagation, magnetic anisotropy, and magnetization reversal [4–9]. However, electric-field control of those magnetic behaviors are far from enough to fully utilize the CME effect. Exchange bias (EB) is an important magnetic property of a coupled antiferromagnetic (AFM)-ferromagnetic (FM) system that occurs due to magnetic interface effects [10]. This effect is widely used in magnetic random access memory cells or giant magnetoresistive read-heads [11]. However, there are few reports about the electric controlled EB with strain-mediated mechanism, which would have promising application in low-power information storage [12,13].

Since the discovery of EB in Co/CoO nanostructure by Meiklejohn and Bean in 1956, this interesting magnetic effect has been extensively studied in various systems containing ferromagnetic, antiferromagnetic, spin-glass and disordered magnetic components [10,14–19]. Recently, large EB effects have been observed

ABSTRACT

We investigate the exchange bias effect in a $Mn_{50,12}Ni_{39,31}Sn_{10,57}/0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ laminate. The exchange bias field of this ferromagnetic shape memory alloy decreases remarkably with the application of electric field, showing an interesting magnetoelectric effect. Using this property, the magnetization sign can be switched in this laminate by electric field without the aid of bias magnetic field. The origin of electric-field control of exchange bias effect is discussed.

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in some Ni–Mn based ferromagnetic shape memory alloys (FSMAs) due to the coexistence of FM and AFM exchange interaction in the martensitic phase [20-26]. In these magnetic functional alloys, the magnetic field can drive the martensitic transformation or the arrangement of martensite variants, leading to the large magnetic-field-induced strain [27]. As a converse effect, the strain can affect the structure of FSMAs as well, which leads to the change of magnetic properties due to the coupling between the structure and magnetism [28]. According to our previous results, in the high-Mn content $Mn_{50}Ni_{40-x}Sn_{10+x}$ (x = 0, 0.5, and 1) alloys, the excess Mn atoms would occupy both Ni and Sn sites and result in the enhancement of AFM exchange interaction, which gives rise to a large exchange bias field (H_E) [26]. In this letter, we report on the electric-controlled EB effect in a Mn-Ni-Sn FSMA/0.7Pb(Mg_{1/} ₃Nb_{2/3})O₃-0.3PbTiO₃ (PMN-PT) laminate. Interestingly, with the application of an electric field, not only the disappearance of EB effect but also the magnetization reversal without bias magnetic field can be observed in this bilayer structure.

2. Experiment

Mn–Ni–Sn ribbons were prepared by melt spinning the as-cast alloys and the average elemental chemical composition of the ribbons was determined as Mn_{50.12}Ni_{39.31}. Sn_{10.57} (MNS), which was examined by the X-ray energy dispersive spectroscopy (EDS) microanalysis. The cubic L2₁ structure of the MNS ribbon was verified by powder X-ray diffraction using Cu Kα radiation at room temperature. The MNS/PMN–PT laminate was prepared by bonding the (001) PMN–PT single crystal with MNS ribbon by epoxy bonder. Here the commercially supplied PMN–PT single crystal was $\langle 011 \rangle$ poled, with the dimensions of $7\langle 001 \rangle^L \times 5\langle \bar{1} \ \bar{1} \rangle^W \times 0.5\langle 011 \rangle^T$ mm³, and coated with Au electrodes as a means of applying ac voltage. The piezoelectric coefficient ($d_{31,p}$) was measured to be -1400 pC/N. Magnetization measurements were carried out using a

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Fig. 1. ZFC and FC thermomagnetic curves for the MNS ribbon under a magnetic field of 100 Oe.

superconducting quantum interference device magnetometer (SQUID, Quantum Design) in a temperature range of 4–320 K with the magnetic field up to 10 kOe. For the thermomagnetic measurement, the zero field cooled (ZFC) and field cooled (FC) processes were performed by cooling the sample from 320 K to 4 K in zero magnetic field and a field of 10 kOe, respectively. The magnetization hysteresis (*M*–*H*) loops of the sample were measured after FC process under the magnetic field of 5 kOe (or -5 kOe) with or without a dc electric field (E_{dc}) applied on the PMN–PT substrate.

3. Result and discussions

Fig. 1 shows the temperature dependence of ZFC and FC magnetization curves for the MNS ribbon. With decreasing temperature, the paramagnetic–ferromagnetic transition occurs at the Curie temperature of the austenite (T_c^A) of about 280 K. Then a sudden decrease of magnetization is observed around 209 K, corresponding to the transition from austenite to martensite. Further decreasing the temperature leads to a FM transition of the martensitic phase. Below the Curie temperature of the martensite (T_c^M), a splitting is observed between the ZFC and FC curves around 112 K and becomes much more pronounced with the decrease of temperature, suggesting the coexistence of AFM and FM exchange interaction in the martensite [21–25].

Fig. 2(a) shows the schematic diagram of the MNS/PMN-PT laminate. In order to study the electric field manipulation of magnetization, E_{dc} is applied along the thickness direction of the

PMN-PT substrate. After FC process, we measure the *M*-H loops for the MNS/PMN-PT laminate at 4 K with E_{dc} = 0 and 4 kV/cm, which are shown in Fig. 2(b). At zero electric field, the magnetization curve shifts completely to the negative field axis with a considerably large H_E of 1086 Oe. Here the value of H_E is calculated as $H_E = -(H_1 + H_2)/2$, where H_1 and H_2 are the left and right coercive fields, respectively. By applying an electric field of 4 kV/cm, a noticeable reduction of H_E with a value of 1059 Oe is observed. It is known that a gradual degradation of H_F upon cycling of the pinned ferromagnet through consecutive hysteresis loops is usually observed in some EB systems, which is termed as training effect [13]. In order to rule out this possibility, we switch off the electric field and measure the *M*-*H* loop for the third time. As shown in Fig. 2(b), H_F of the MNS ribbon restores its initial value instead of further decreasing, suggesting that the modification of EB stems from the electric field.

Fig. 3(a)–(d) shows the E_{dc} dependence of M–H loops for the laminate at different temperatures (10, 30, 45, and 67 K) measured after FC process with a cooling field of 5 kOe. Remarkable changes of H_E under the manipulation of E_{dc} can be seen from Fig. 3. At 10 K, H_E of the sample decreases from 952 to 907 Oe with the application of E_{dc} = 4 kV/cm, which is shown in Fig. 3(a). In the case of 30 K, a much more pronounced effect of E_{dc} control of EB is observed, exhibiting a remarkable downward shift of H_E from 480 to 205 Oe under E_{dc} = 4 kV/cm, as shown in Fig. 3(b). With increasing temperature at 45 K (Fig. 3(c)), M-H loop gradually shifts to the positive axis while H_E shows rapidly reduction from 200 to 120 Oe with E_{dc} . As shown in Fig. 3(d), H_E of 67 K is 16 Oe with zero electric field. However, by applying E_{dc} = 4 kV/cm on PMN–PT substrate, a symmetric *M*-*H* loop without EB is observed, suggesting the FM regions are no longer pinned and the FM domains begin to rotate [17,26].

The shift of M-H loops by E_{dc} reveals the electric field controlled EB effect in the laminate, which can be understood in terms of the strain-mediated magnetic properties of MNS ribbon. According to the Meiklejohn Bean expression [10,14]:

$$\mu_0 H_E = -\frac{JS_{\rm FM}S_{\rm AFM}\cos\phi}{M_{\rm FM}t_{\rm FM}},\tag{1}$$

where ϕ is the angle between the interface magnetizations of the FM layer and the AFM pinning layer, *J* is the interface exchange constant between the FM and AFM interface magnetizations *S*_{FM} and *S*_{AFM}, *t*_{FM} and *M*_{FM} are the thickness and the saturation magnetization of the FM layer, respectively. In the MNS/PMN–PT laminate, with the application of *E*_{*dc*} on the piezoelectric substrate, the transferred strain would modify the crystallographic configuration of the



Fig. 2. (a) Schematic diagram of the MNS/PMN-PT laminate. (b) M-H loops for the MNS/PMN-PT laminate at 4 K with and without Edc.

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