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Voltage-controllable magnetic behavior in PZT/NiFe/CoFe nanocomposites

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Abstract

Recently, the electric field induced manipulation of magnetic properties has become one of the most interesting research topics because of its promising applications in magnetoelectronics, such as future logic elements and electric–magnetic memories. In this paper, the NiFe/CoFe ferromagnetic nanostructured thin films were directly deposited on the ferroelectric PZT substrate, which was polarized parallel to the thickness or along the surface. In these samples, the stress originating from the piezoelectric layer is transferred into the magnetostrictive layer, which induces an elastic strain and results in a change in magnetization, thanks to the converse magnetoelectric effect. By analyzing the magnetization of the nanocomposite under a fixed magnetic field up to 5×10^3 Oe at different voltages applied to the ferromagnetic substrate, we can investigate the voltage-controllable magnetization in this composite. The obtained results show that the magnetization of the NiFe/CoFe film changes around 6% at the low voltage of 60 V and up to two times larger at 800 V in an external magnetic field of 50 Oe. The use of voltage also allows the reversible adjustment of the magnetization orientation in ferromagnetic layers.

Keywords: ferroelectrics, lead zirconate titanate PZT, ferromagnetics, converse magnetoelectric effect

Classification numbers: 4.10, 4.11, 5.02, 5.11

1. Introduction

Multiferroics are the materials that can be either electrically polarized or magnetized. A number of research projects predict that these properties are predominant in two-phase composites consisting of separate piezoelectric and magnetostrictive phases ([1–3] and reference therein). In multiferroics, the direct magnetoelectric effect (DME) is characterized by the polarization response P to an external magnetic field H , while the converse magnetoelectric effect (CME) is exhibited by the magnetization M response to an applied electric field E . On the other hand, the deformation of the magnetostrictive phase causes polarization of the piezoelectric phase in the composite and *vice versa*. Promising scenarios relating to applications of the ME effect are magnetic field sensors, logic elements and electric write-magnetic read memories [2–4].

In recent years, various kinds of multiferroics have been investigated, including single-phase perovskites, such as BiFeO_3 , BaMnF_4 , to bulk materials composed of both ferroelectric and ferromagnetic phases and layered materials consisting of ferromagnetic and ferroelectric layers, such as $\text{BaTiO}_3\text{--CoFe}_2\text{O}_4$, $\text{BaTiO}_3\text{--NiFe}_2\text{O}_4$, $\text{PZT--CoFe}_2\text{O}_4$ and PZT/Tefernol-D [5–9]. For multi-phase materials, to achieve better coupling in the properties, most reported work has focused on the laminate structure of ferromagnetic materials with a high magnetostriction coefficient, such as Tefernol-D, Fe-rich based ribbon, ferrites and common piezoelectric materials, such as lead zirconate titanate (PZT) and later polyvinylidene fluoride (PVDF) [10]. However, recent work has indicated that one can use transition metals and their alloys as the magnetostrictive phase to improve the performance of the composites [11–13]. The primary advantages of such structures are to accomplish low leakage



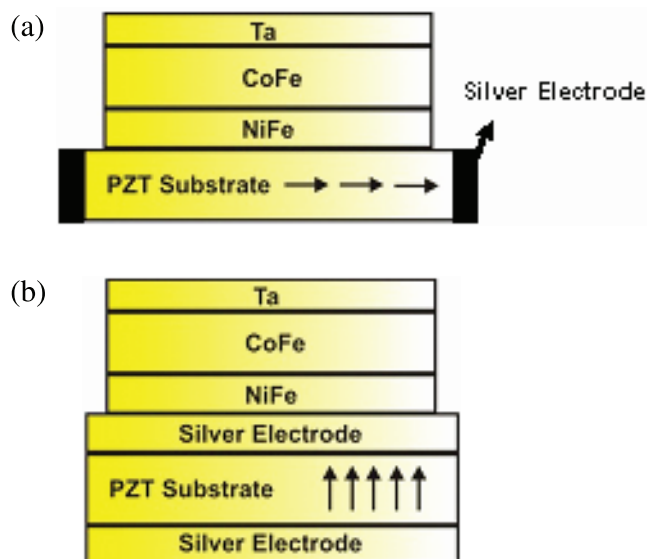


Figure 1. Schematic diagram of PZT/NiFe/CoFe composites: (a) transverse and (b) longitudinal polarized PZT.

current and a high degree of polarization compared to oxide ferromagnets [14].

In this paper, the properties of bilayer ferromagnetic thin films, NiFe/CoFe, deposited on commercial PZT substrates were studied. The voltage-controllable magnetization of this composite was investigated by analyzing the magnetization of the nanocomposite under external magnetic field at different applied voltages. The effect of the polarization direction of the PZT substrate on magnetization behavior will be discussed.

2. Experimental

The composite structures, consisting of two ferromagnetic magnetostrictive layers NiFe and CoFe, are directly grown on the piezoelectric PZT substrates by means of the rf magnetron sputtering 2000-F (AJA International). The commercial PZT substrates, which were polarized either parallel to the thickness or along the surface, have been used. The main chamber was vacuumed to a base pressure of less than 2×10^{-7} Torr before sputtering.

The ferromagnetic layers of NiFe and CoFe were deposited in sequence on the PZT substrates at room temperature and in an Argon pressure of 2.2×10^{-3} Torr using rf power of 50 W. The thickness of the NiFe and CoFe layers was changed by varying the sputtering time of each layer, of about 100 nm. Finally, a Ta thin layer was sputtered onto the film surface to protect the ferromagnetic layers from oxidization. For the CME measurement, the electrodes were made by using silver adhesive glue in the geometry shown in figure 1. The composite films, deposited on the transverse and longitudinal polarized PZT substrates, were denoted as samples P1 and P2, respectively.

The crystallographic structure of the composite films was investigated by using an x-ray diffractometer (XRD) Advance 8 (Bruker) with a $\text{CuK}\alpha$ wavelength. The surface morphology was analyzed by using a scanning electron microscope (SEM) S-3400N (Hitachi). The voltage-controllable magnetization behavior of these composites was measured by using a vibrating sample magnetometer (VSM) 7400 (Lakeshore)

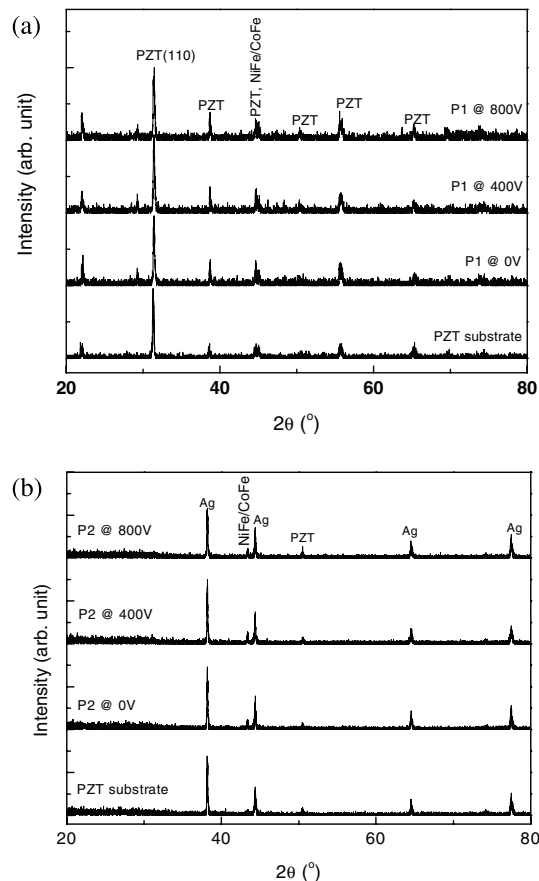


Figure 2. XRD patterns of (a) sample P1 and (b) sample P2 measured at different applied voltages.

with the magnetic field aligned parallel to the sample plane. In this measurement, the voltage applied to the PZT substrate was also varied from -800 V to $+800$ V by using a voltage amplifier.

3. Results and discussion

The XRD patterns, measured at different applied voltages, of PZT/NiFe/CoFe composites are presented in figure 2. The XRD pattern of PZT substrate is also included for comparison. For all samples, most of the diffraction peaks in the XRD patterns can be indexed for the perovskite structure of PZT. For sample P1, there is a main PZT peak at 2θ of 31.3° , corresponding to the (110) orientation (see figure 2(a)). Other PZT and NiFe/CoFe reflections appear at 2θ of about 44 – 45° . For sample P2, besides Ag and small PZT reflections, the peak at 44° belongs to NiFe/CoFe (see figure 2(b)). The NiFe/CoFe reflection is clearly observed in this sample. It is also noted that there is a small shift in PZT peak to a high angle region with increasing applied voltage. This can be explained by the small change in the lattice parameters of the PZT unit cell that is induced by an electrical field.

The SEM images of P1 and P2 film surfaces are illustrated in figure 3. It is clear that the ferromagnetic layers follow the morphology of the PZT substrate. The grain sizes are in the range of a few tens of μm for sample P1 to $10\mu\text{m}$ for sample P2 with high surface roughness. For sample P2 with longitudinal polarization, the surface is somehow finer

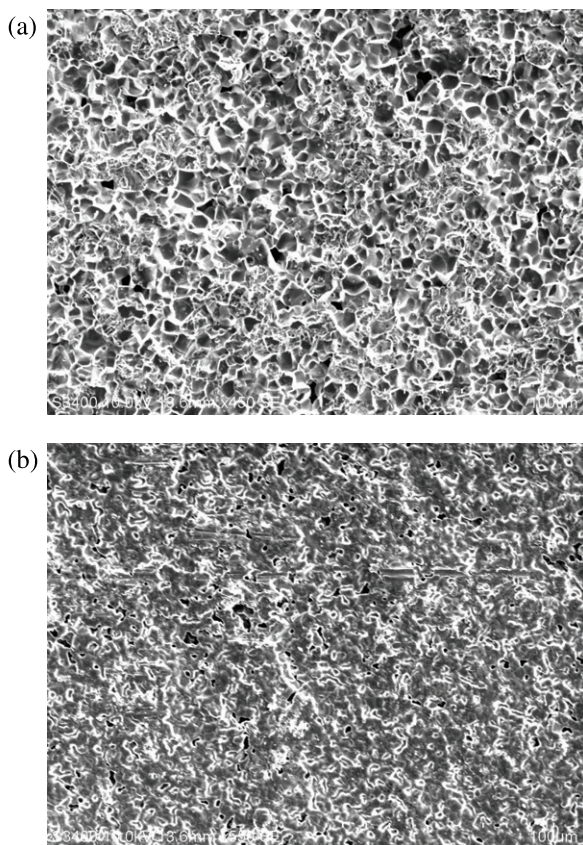


Figure 3. SEM image of PZT/NiFe/CoFe structures: (a) sample P1 and (b) sample P2.

because of a thin silver layer covering the top of the substrate (figure 3(b)).

Figure 4 shows the magnetic hysteresis loops measured in magnetic fields applied parallel, 45° and perpendicular to the sample plane at room temperature for samples P1 and P2. The in-plane magnetic anisotropy predominates in both samples due to the contribution of the magnetic anisotropy from NiFe/CoFe layers. In contrast to sample P1, the magnetization curve in the film plane of sample P2 is close to the saturated state.

For this type of composite, imposing a strain on a magnetic layer by a mechanical stress may alter the magnetization as well as the shape of the hysteresis loops. Using an electric voltage/field, one can induce a stress in a piezoelectric substrate and consequently in a ferromagnetic layer. This means that by this method the voltage-controllable magnetization can be achieved. For example, the sample P1 shows an obvious electric field influence on the magnetic hysteresis loops while applying the electric field along the sample plane. The coercivity increases from 63 Oe to 80 Oe at a voltage of 600 V (not presented here) and the ferromagnetic layers appear to be magnetically harder when the substrate is polarized by an electric field. The coercivity can be manipulated by 21% in this sample, which is larger than the recently reported value in Ni/BaTiO₃ hybrid structures [15].

For more detailed investigations, the voltage dependence of magnetization has been analyzed for samples P1 and P2 at various magnetic fields from 50 to 5000 Oe. As depicted in figure 5, the change in magnetization is virtually linear with changing voltage but very much depends on the polarization

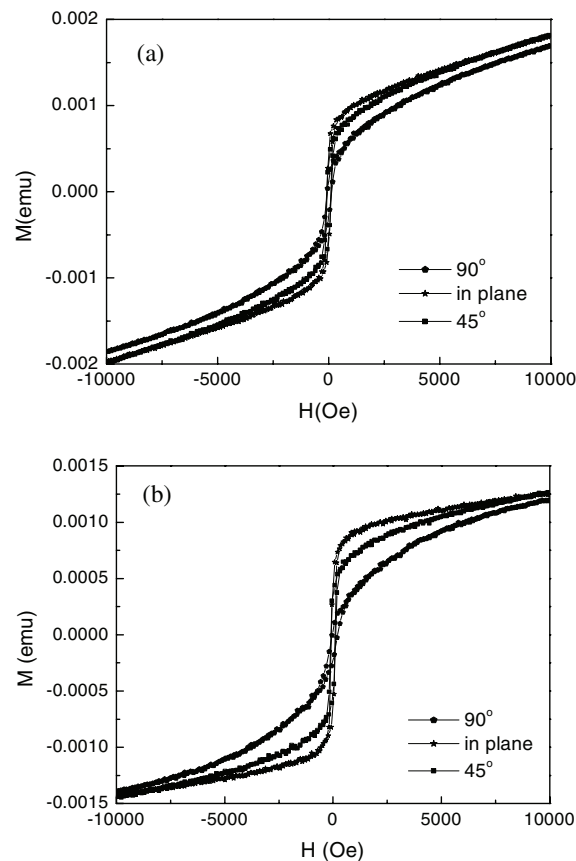


Figure 4. Magnetic hysteresis loops of (a) sample P1 and (b) sample P2.

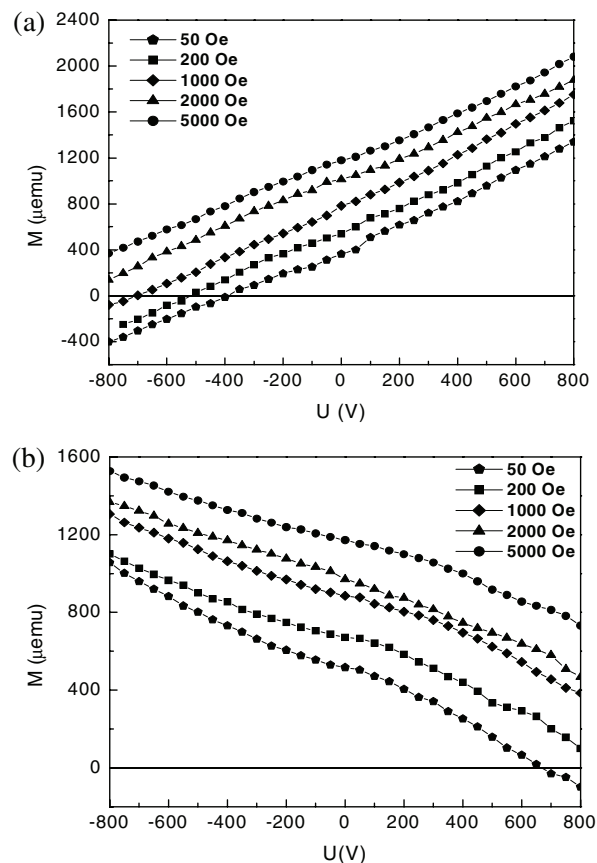


Figure 5. The change in magnetization measured at various magnetic fields for (a) sample P1 and (b) sample P2.

direction of the PZT substrate. For sample P1 having a transversely polarized PZT substrate, the magnetization increases when changing the voltage from -800 V to $+800$ V (see figure 5(a)). In contrast, the magnetization decreases for sample P2 having a longitudinally polarized PZT substrate (figure 5(b)).

This demonstrates that the elastic stress transferred from the PZT substrate into the ferromagnetic NiFe/CoFe layer is the driving force here. In sample P1, the magnetic field and the electric field are parallel to the sample plane, so the stress is tensile. This resulted in increasing magnetization with increasing applied voltage. For sample P2, the induced stress is compressed in the film plane, which resulted in decreasing magnetization. The relative change in magnetization of the NiFe/CoFe thin film is significant, around 6% at the low voltage of 60 V and up to 100% and 200% at 800 V in an external magnetic field of 50 Oe for samples P2 and P1, respectively.

Moreover, there exists a reversible magnetization with changing applied voltage. For sample P1, this process occurs at 400 V in a magnetic field of 50 Oe that is much smaller than the value of 650 V for sample P2. This is evidence that one can manipulate the magnetization of the ferromagnetics by using a suitable voltage.

4. Conclusions

The hybrid structures of ferroelectric–ferromagnetic PZT/NiFe/CoFe have been fabricated and analyzed under the effect of external magnetic and electric fields. The magnetization behavior strongly depends on the polarization direction of the PZT substrate. It is worth noting that the relative change in magnetization of the ferromagnetic layer is very large, up to 200%. Furthermore, we can manipulate the reversible magnetization process in this structure by using a suitable voltage. This result is promising from a practical point of view, such as in magnetoelectronic devices.

A study of the thickness effect of ferromagnetic layers in voltage-controllable magnetization behaviors is under analysis.

Acknowledgments

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