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Large converse magnetoelectric effect in ferromagnetic shape memory alloy $Ni_{49}Fe_{18}Ga_{27}Co_6$ and $Pb(Zr_{0.52}Ti_{0.48})O_3$ laminates

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ABSTRACT

Ferromagnetic shape memory alloy $Ni_{49}Fe_{18}Ga_{27}Co_6$ was used in a laminated composite with $Pb(Zr_{0.52}Ti_{0.48})O_3$ for studying the converse magnetoelectric (CME) effect. The CME properties with the frequency of driving electric voltage, bias magnetic field, and dc bias electric voltages were investigated in detail. A large value of α_B of 1.05 G/V at the resonance frequency 89 kHz is observed under a low optimum H_{bias} of 100 Oe. The maximum values of α_B at 1 kHz can be tuned from 8.7 to 25 mG/V by increasing dc voltage bias from −100 to 100 V, exhibiting a giant tunable range in α_B values. This composite exhibits promising applications in electrically controlled magnetic force devices.

1. Introduction

The magnetoelectric (ME) materials have received considerable attention during the past decades due to their potential applications in information storage, sensors and microwave systems [1–5]. Compared with the weak coupling in most of single-phase multiferroics, orders of magnitude large ME coefficients can be achieved in some laminate composites, which are comprised of piezoelectric and magnetostrictive materials [5–10]. ME effects in these laminated composites originate from similar mechanism of strain mediated coupling between the magnetic and electric subsystems and can be generally classified into direct ME effect (DME) and converse ME (CME) effect [3]. The DME effect is an electric polarization response of a material to an applied magnetic field, while the CME effect is a magnetic response of a material to an applied electric field. The CME effect, however, is not studied as widely as the DME effect, especially in multiphase composites. The working principle for CME effect in these composites is as follows: when an ac electric field is applied to the piezoelectric materials, the reverse piezoelectric effect would lead to a mechanical force; then this force is transferred to the magnetic layer and induces the variation of magnetization due to the piezomagnetic effect [11–14]. In recent years, Terfenol-D has been the most widely used alloy in studying ME effect for its excellent magnetostrictive property [5,11,12]. However, the intrinsic brittleness and high-cost of Terfenol-D has stimulated the researchers to use other alloys as magnetic layer in ME laminated composites [8,13–15].

Ferromagnetic shape memory alloys (FSMAs) are a kind of multifunctional materials that show ferromagnetic and shape memory effect simultaneously. Since the magnetic field can drive the martensitic transformation (MT) or the arrangement of martensite variants, large magnetic-field-induced strain (MFIS) can be observed in these alloys [16]. As a converse effect, the stress can affect the structure of FSMAs as well, which can lead to the change of magnetization due to the coupling between the structure and magnetism. Accordingly, an alternative mechanism for obtaining large CME effect in the FSMAs/piezoelectric laminates was reported, in which the stress induced MT or rearrangement of martensite variants is used to gain the magnetic changes in FSMAs [13]. Giant resonance CME coefficients (α_B) of 2.91 G/V and 18.6 G/V were observed in the FSMA Ni–Mn–Co–Sn/PZT and Ni–Mn–Ga/$0.7Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.3PbTiO_3$ (PMN-PT) single crystal laminates [8,13], respectively, indicating that FSMAs are the promising magnetic layers in ME composites. Recently, Ni–Fe–Ga alloys have been developed as potential FSMAs [17,18]. However, the magnitude of the MFIS in Ni–Fe–Ga system is small at room temperature due to the reduced MAE resulting from the low Curie temperature of these alloys [19]. Experimentally, Morito et al. reported that both the Curie temperature and the magnetocrystalline anisotropy energy of Ni–Fe–Ga alloys can be increased by adding Co elements [20,21]. As a result, a large MFIS of 0.7% was observed in $Ni_{52}Fe_{18}Ga_{27}Co_3$ alloy [21]. Moreover, an enhanced

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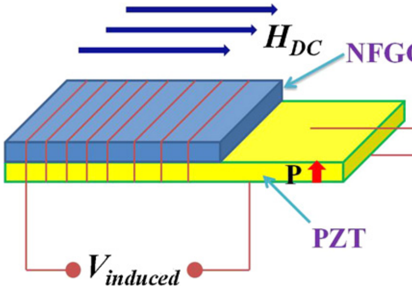

Fig. 1. Schematic diagram of the NFGC/PZT CME laminated composite.

MFIS of 8.5% could be obtained in $Ni_{49}Fe_{18}Ga_{27}Co_6$ (NFGC) alloy with a static compressive stress [20]. Owing to the strong coupling between elastic and magnetic domains in NFGC alloy, large changes in magnetization could be anticipated in the process of twin-variant motion driven by an applied magnetic field or mechanical stress. In this paper, we investigated the CME effect in a NFGC/$Pb(Zr_{0.52}Ti_{0.48})O_3$ (PZT) laminate. Interestingly, the values of α_B for this laminate can be tuned in a large scale by applying different dc electric voltage bias.

2. Experimental details

Polycrystalline NFGC alloy was prepared by arc-melting method. The as-cast alloy was sealed in quartz tube and annealed at 1420 K for two days to homogenize and then quenched in ice water. The annealed alloy was re-heated at 873 K for two days to obtain large antiphase domain and then kept at 673 K for one day followed by furnace cooling [20,21]. The PZT plate was commercially supplied with a piezoelectric coefficient of 580 pC/N and polarized along the thickness direction. The dimensions of NFGC and PZT are L8.50 mm × W5.30 mm × T0.50 mm and L14.50 mm × W5.30 mm × T0.50 mm, respectively. The schematic diagram of the NFGC/PZT composite is shown in Fig. 1. It was prepared by bonding the NFGC alloy with PZT ceramic chip by epoxy bonder, which was placed in a dc bias magnetic field (H_{bias}) and designed to operate in the L-T ME coupling mode (longitudinal magnetization and transverse polarization). The magnetostriction of the NFGC alloy was measured by standard strain gauge method with magnetic field parallel to the longitudinal axis. A search coil with 35 turns of Cu wire was wrapped around the composite to measure the induced magnetic induction (B_{ind}). An applied sine ac electric voltage (V_{ac}) at a prescribed frequency (f) was supplied by an arbitrary function generator (NF 1973) and a power-supply amplifier (NF HAS 4051) to excite the PZT plate in the NFGC/PZT composite. The change in magnetic flux associated with the NFGC plate was measured by a lock-in amplifier (SRS Inc., SR810).

3. Results and discussions

Fig. 2 shows the frequency dependence of the α_B for the composite under an optimum H_{bias} of 100 Oe and with applied V_{ac} of 10 V. The value of α_B in NFGC/PZT composite is defined as magnetic response to an applied ac electric voltage, $\alpha_B = dB/dV$ [8,11,12]. Two major and three minor peaks of α_B are observed within the frequency of 1–104 kHz. The peak values are 0.07, 0.40, 0.16, 0.22, and 1.05 G/V at the frequency of 8, 25, 43, 74, and 89 kHz, respectively, corresponding to the electromechanical resonance frequencies [8,11,12]. The maximum value of α_B (1.05 G/V) is larger than that in Terfenol-D based laminates [11,22] and comparable with that in FSMA Ni–Co–Mn–Sn and PZT bilayer composite [13]. The magnetic field dependence of the magnetostriction for the NFGC alloy is shown in the inset of Fig. 2. It is obvious that the value of magnetostriction is less than that of Terfenol-D and reaches only 24 ppm at a low magnetic field of 200 Oe, which would be attributed to the domain wall motion of the NFGC alloy. As mentioned above, the CME effect is related to the reverse process of the magneto-mechanical response, that is, the effect of the stress on the magnetization. It is reported that the critical stress for the motion of the martensite variants of the NFGC alloy is quite small [19]. In the case of NFGC, besides the domain wall motion, the stress transferred from the PZT plate would lead to the motion of the twin boundary as well, giving rise to the considerable change of magnetization due to the coupling between the structural and magnetic properties. Therefore, both the motion of the magnetic domain wall and the twin boundary give rise to the variation of the magnetization of NFGC. It is reported that laminate composite operated in L-T mode can be analyzed using a quasistatic model, in which the boundary condition is described as [5,11]

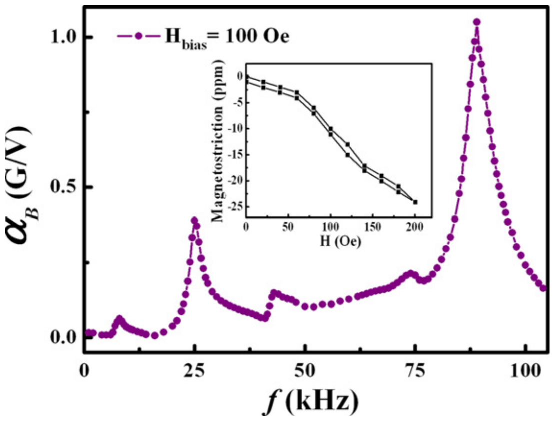

Fig. 2. The frequency response of CME coefficient under an applied ac electric voltage of 10 V (peak) and a H_{bias} of 100 Oe. Inset: The magnetic field dependence of magnetostriction for NFGC plate in the longitudinal direction.

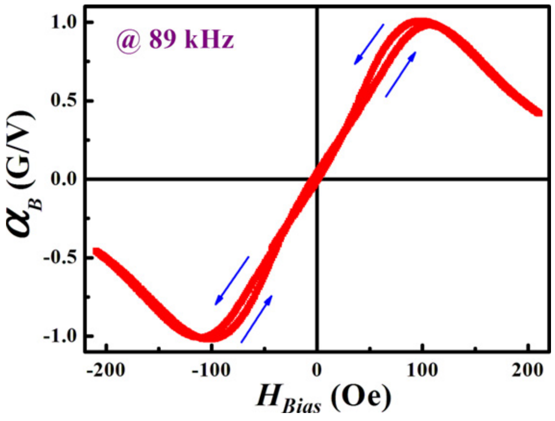

Fig. 3. The H_{bias} dependence of CME coefficient at the resonance frequency of 89 kHz.

$$T_{1,p}2t_p = -T_{3,m}t_m \tag{1}$$

$$S_{1,p} = S_{3,m} \tag{2}$$

where $T_{1,p}$ and $S_{1,p}$ are the stress and strain of the PZT along the length direction, while $T_{3,m}$ and $S_{3,m}$ are the stress and strain of the NFGC along the length direction, t_p and t_m are the thickness of the PZT and NFGC, respectively. Eventually, the converse magnetoelectric coefficient (α_B) of the composite can be expressed as

$$\alpha_B = \left|\frac{dB_3}{dV_3}\right| = \left|\frac{2d_{31,p}d_{33,m}}{s_{11}^E t_m + 2s_{33}^H t_p}\right| \tag{3}$$

where s_{11}^E and s_{33}^H are the elastic compliances of the piezoelectric and magnetostrictive layers, respectively, while the values of $d_{31,p}$, $d_{33,m}$ are the piezoelectric coefficient and piezomagnetic coefficient. According to Eq. (3), large α_B in NFGC/PZT would be ascribed to the cooperation of large $d_{31,p}$ in piezoelectric layer and large $d_{33,m}$ in piezomagnetic phase.

Fig. 3 shows the α_B as a function of H_{bias} at the resonance frequency of 89 kHz. As magnetic field increases, α_B increases initially up to the maximum of 1.02 G/V at 100 Oe and then decreases under higher field. The optimum H_{bias} value of present laminated

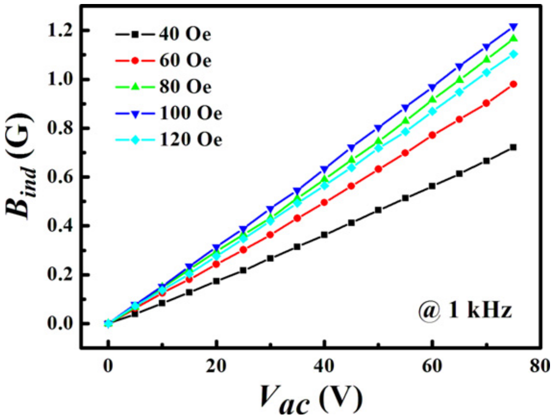

Fig. 4. B_{ind} as a function of V_{ac} (peak) with different H_{bias} at the frequency of 1 kHz.

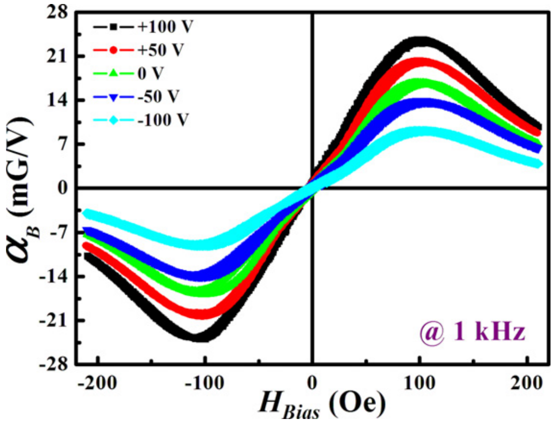

Fig. 5. The CME coefficient dependence of H_{bias} with different dc voltage biases at the frequency of 1 kHz.

composite is 100 Oe, which is remarkably less than that of Terfenol-D/PMN-PT and Ni–Co–Mn–Sn/PZT laminates [11,13,22]. As we know, the magnetization change of NFGC can be induced by not only magnetic field but also stress. Therefore, the behavior of α_B in NFGC/PMN-PT laminated composite is determined by the cooperation of H_{bias} and stress which is transferred from the piezoelectric layer. The peak value in α_B-H_{bias} curve means that at the optimum magnetic field of 100 Oe, the H_{bias}-and-stress-induced change of magnetization is the largest. Further increasing of H_{bias} would lead to the decrease of magnetic change. Moreover, According to the results of Fig. 3, it is obvious that α_B depends significantly on the frequency of driving electric field and is highly sensitive to the external dc magnetic bias, so present composite with large CME effect could be used to detect small changes of the dc magnetic field [5,23].

The magnetic induction (B_{ind}) as a function of applied ac voltage (V_{ac}) with different H_{bias} at the frequency of 1 kHz is plotted in Fig. 4. It is worth noting that B_{ind} has good linear responses to the applied ac voltage at the range of 0–75 V (peak) for all H_{bias}, which is consistent with the earlier reports [8,13]. Obviously, α_B increases with the increasing H_{bias} and reaches the maximum of 17 mG/V at 100 Oe. Further increasing H_{bias} (120 Oe) leads to the decrease of α_B. Similar behavior of α_B at optimum H_{bias} = 100 Oe has been observed at resonance frequency 89 kHz as shown in Fig. 3. Importantly, even if the ac driving frequency is 1 kHz, well below the resonance frequency, significant CME responses can also be observed at low H_{bias}, indicating high sensitivity of CME effect in this laminated composite which is very important from the point of application view.

As mentioned above, the MFIS of NFGC alloy is enhanced under a static compressive stress, which can be produced by applying a dc electric voltage bias on the piezoelectric layer in the case of NFGC/PMN-PT laminated composite. Fig. 5 shows the H_{bias} dependence of the α_B with dc electric voltage bias varying from −100 V to 100 V. The excitation ac voltage is 10 V (peak) at the frequency of 1 kHz. All the curves show similar behaviors with the variation of H_{bias} and peak at the same optimum field of 100 Oe despite of different dc voltage bias. It is worth noting that the CME effect is significantly modified by applying dc electric voltage on PZT plate, which is enhanced by positive voltage and reduced by negative one. As we know, the applied dc bias voltage would generate a static stress on the NFGC plate, and then affect the rearrangement of its twin boundary. With the positive voltage, the additional compressive stress facilitates the twin boundary motion [20], leading to a larger magnetization variation of the NFGC plate. With the dc voltage bias increasing from 0 to 100 V, the maximum value of the α_B increases from 17 to 25 mG/V, as a result of the enhanced CME effect, as shown in Fig. 5. In contrast, as the voltage varies from 0 to −100 V, the maximum value of α_B decreases from 17 to 8.7 mG/V, due to the fact that the twin boundary mobility would be weakened by the tensile stress load on the magnetic crystal. Therefore, the α_B of present composite can be effectively tuned by applying dc bias voltage on the piezoelectric layer [24]. As the bias voltage increases from −100 to 100 V, the maximum value of α_B changes by more than 180%, exhibiting a giant tunable range in α_B values.

4. Conclusions

In summary, a large CME effect has been observed in a bilayer composite bonding a FSMA NFGC layer and a PZT plate polarized in the thickness direction. The effects of frequency, H_{bias}, and dc bias electric voltage on the CME coefficients of the composite are investigated. A large value of α_B of 1.05 G/V at the resonance frequency 89 kHz is obtained under a low optimum H_{bias} of 100 Oe. An excellent linear relationship between B_{ind} and V_{ac} was observed in the range of 0–75 V (peak) for all H_{bias}. The CME effect can be effectively tuned by applying dc electric voltage bias on PZT plate. The maximum values of α_B at 1 kHz can be tuned from 8.7 to 25 mG/V by varying voltage bias from −100 to 100 V. These results demonstrate that this laminated composite possesses a large CME effect under low H_{bias} and tunable maximum values of the α_B under different dc voltage bias, suggesting its promising potential in the application of data storage, actuators, and sensors.

Acknowledgments

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