

Effects of alternating magnetic field on magnetoelectricity of sputtered TbFe₂/PZT/TbFe₂ laminate composite

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Both magnitude and frequency of alternating magnetic field (H_{ac}) affect the magnetoelectric (ME) effect in a sputtered TbFe₂/PZT/TbFe₂ laminate composite. With the increase in H_{ac} from 5 to 20 Oe, the magnitude of ME susceptibility (the ratio of induced electric polarization to H_{ac}) is decreased indicating an insensitive polarization to the variation of H_{ac} in this regime. When the frequency of H_{ac} is lower than 2.5 kHz, the ME susceptibility is increased with increasing bias magnetic field (H_{dc}) up to 280 Oe. With further increases in H_{dc} , the ME susceptibility drops to a saturation value. By contrast, the ME susceptibility in the case of H_{ac} beyond 2.5 kHz is monotonically increased by the increase in H_{dc} up to 4500 Oe. At 2.5 kHz, the ME effect has a linear variation with the H_{dc} ranging from 800 Oe to 4500 Oe.

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1. Introduction

Magnetoelectric (ME) composites comprise the piezoelectric materials (e.g. PbZr_{1-x}Ti_xO₃ or PZT, BaTiO₃, polyvinylidene fluoride or PVDF) and magnetostrictive materials (e.g. soft ferrites, Fe, Co, Ni, Tb_{1-x}Dy_xFe₂ or Terfenol-D) [1,2]. In an applied magnetic field, the mechanical stress originated in the magnetoactive phase is transferred to the electroactive phase and the electric field is induced as a result of the piezoelectric effect. Likewise, the mechanical stress in the electroactive phase upon the application of electric field induces the magnetic field [1]. The conversion between the electric and magnetic fields in ME composites is of interest for both theoretical and technological aspects. Compared with the particulate composites [3-5], the laminate composites generally exhibit higher ME effect due partly to less mechanical defects and undesirable phases [1]. In ME sandwich structures, magnetoactive layers on both sides of an electroactive disk can be achieved by several methods including sputtering, aerosol deposition as well as epoxy bonding [2]. In order to utilize positive magnetostriction, bonded terfenol-D is commonly used [6,7]. Rare earth-transition metal Tb-Fe also exhibits the giant magnetostriction effect but its implementation in micro-electromechanical systems (MEMS) is impeded by its large magneto-crystalline anisotropy [8,9]. To reduce the magnetic fields needed to switch Tb-Fe components and enhance their applications, the sputtered TbFe layers are coupled with soft ferromagnetic films [10,11]. So far, there have been a few publications on incorporating TbFe₂ into ME structures with only a single report on using sputtering [12-14]. In this work, a laminate ME composite

is prepared by sputtering of both sides of a PZT disk. The ME effect is studied as a function of magnitude and frequency of the magnetic field to explore the working characteristics in sensing applications.

2. Experimental

The TbFe₂/PZT/TbFe₂ laminate composite was prepared by rf magnetron sputtering technique. A PZT disk of 6.4 mm in diameter and 0.191 mm in thickness (Piezo Systems Inc. model PSI-5A4E) was used as a substrate with 7.6 cm-diameter TbFe₂ disk (99.9% purity) installed as a sputtering target. Before deposition, the chamber was pumped down to a base pressure below 10⁻⁵ mbar and then Ar gas up to 51 sccm was continuously supplied to the chamber. The rf power was fixed at 300 W during the entire deposition. After the deposition, the sample was examined by scanning electron microscopy (SEM) and its magnetic properties were measured by vibrating sample magnetometry (VSM).

In the ME measurement as illustrated in Fig. 1, the sample was subjected to the alternating magnetic field (H_{ac}) and bias magnetic field (H_{dc}) in the direction parallel to the sample plane. An electromagnet (Walker Scientific Inc. model HV-4H with a power supply) was used to generate the H_{dc} up to 4.5 kOe. A power amplifier (TOA Corporation model A-2120H) was used to amplify the sinusoidal signal from the lock-in amplifier (Stanford Research Systems model SR850) and then supplied to a pair of Helmholtz coils to generate H_{ac} . Both H_{dc} and H_{ac} were measured by a gaussmeter (LakeShore model 455) equipped with a Hall probe (LakeShore model HMMT-

6J04-VR). The electric charge induced by the sample was measured by a charge amplifier (Kistler model 5015A) and the output voltage from the charge amplifier was measured by the lock-in amplifier. The ME susceptibility (α) is defined as

$$\alpha = \frac{P}{H_{ac}} \quad (1)$$

where P represents the electric polarization (charge per unit area).

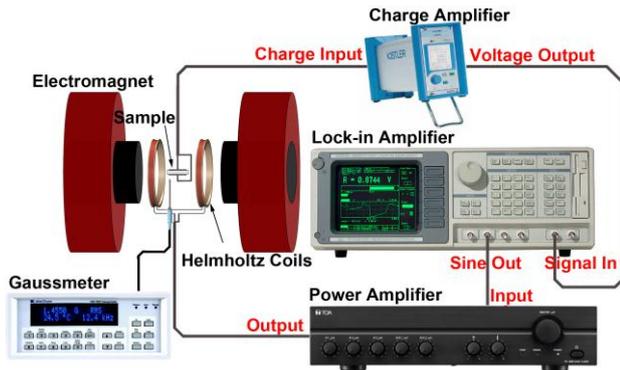


Fig. 1. Schematic diagram of the ME measurement system.

3. Results and discussion

Fig. 2 shows a cross-sectional SEM image of the sputtered TbFe₂ film on the PZT disk. The TbFe₂ appeared as a lighter layer is $2.0 \pm 0.1 \mu\text{m}$ thick. The room temperature magnetic hysteresis loop obtained from the VSM measurement of the TbFe₂/PZT/TbFe₂ composite is shown in Fig. 3. With the magnetic field parallel to the plane of the sample, the magnetoactive TbFe₂ layers contribute to the saturation magnetization of 0.15 emu/g and the remanent magnetization of 0.05 emu/g. The coercive field around 100 Oe is generally higher than those of Tb-Fe alloys in other ratios. In addition to the dependence on the composition, Jiang and co-workers demonstrated that the magnetic behavior of Tb-Fe films were sensitive to the pressure and plane of sputtering [8,9].

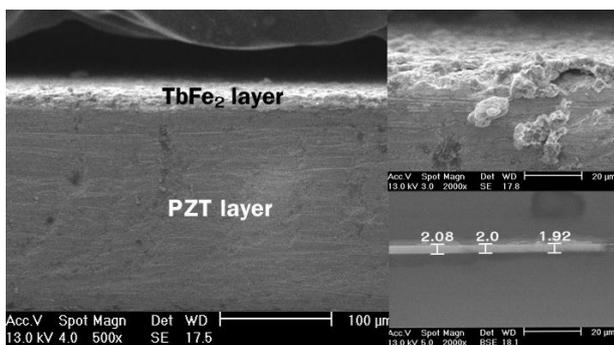


Fig. 2. SEM images of a TbFe₂ layer deposited on PZT surface.

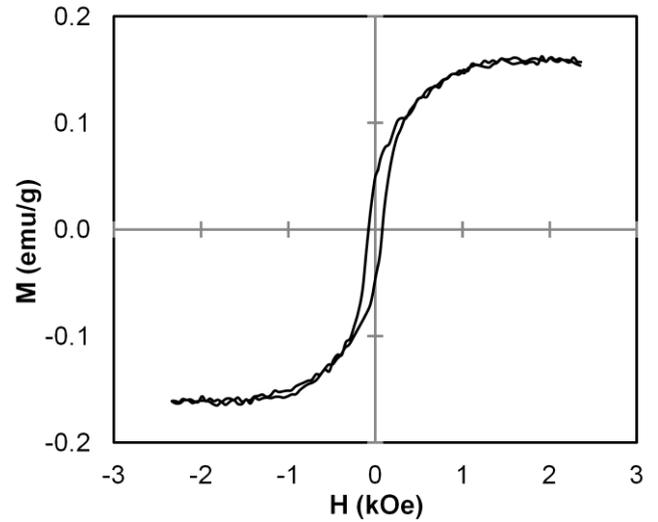


Fig. 3. Room temperature magnetization curve of the trilayered TbFe₂/PZT/TbFe₂ composite.

The strength of electroactive and magnetoactive responses as well as the coupling between these phases, which contribute to the ME effect, can be expressed in terms of the ME susceptibility. In Fig. 4, the ME susceptibility is plotted as a function of H_{dc} . In this particular measurement, H_{ac} of 7 different frequencies far below the resonance were studied at a fixed magnitude of 10 Oe. The ME variation with H_{dc} has a few distinct characteristics depending on the frequency. When the frequency of H_{ac} is lower than 2.5 kHz, the ME susceptibility is increased with the increase of H_{dc} in a low regime and reaches maximum at the value called the characteristic field. The characteristic field around 280 Oe in Fig. 4 is comparable to the external field which causes the rotation of magnetization indicated by the hysteresis loop in Fig. 2. Beyond the characteristic field, the ME susceptibility is gradually reduced with the further increase in H_{dc} before approaching the saturation at high fields. A similar ME response at 1 kHz was previously observed in epoxy bonded TbFe₂/PZT/TbFe₂ composites [13]. In contrast to low frequencies, the ME susceptibility in the case of 3.0 and 3.5 kHz is monotonically increased with increasing H_{dc} . When the frequency of H_{ac} is 2.5 kHz, the characteristic can be considered as an immediate transition between the low and high frequency regimes. Interestingly, the ME susceptibility at this frequency exhibits a linear response to H_{dc} in an extended range from 800 Oe to 4.5 KOe. The characteristics of ME in Fig. 4 can be explained by the magnetostrictive behavior of TbFe₂. The maximum ME effect occurs when the variation in magnetostriction reach the maximum. Further increases in magnetic field are no longer enhance the response in the piezoelectric phase but only reduce the ME coefficient and susceptibility. From the previous works, the interesting features in TbFe₂ films are its approximately linear magnetostriction in response to H_{dc} and high saturation field [8,13]. As evident from this work, the magnetostrictive behavior of TbFe₂ is also influenced by

the frequency of H_{ac} . In Fig. 5, the ME susceptibility is plotted against the frequency of H_{ac} . The increase in ME effect with increasing frequency far below the resonance was similar to the nickel ferrite/PZT multilayers and it can be explained by the variation of electrical conductivity of magnetoactive layers in this frequency regime [15].

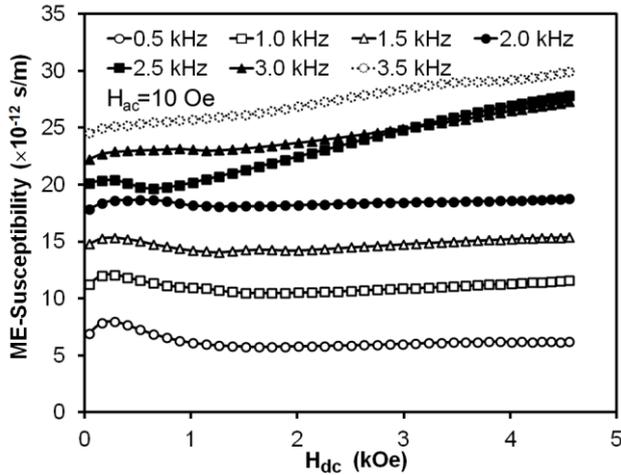


Fig. 4. ME responses of the trilayered $TbFe_2/PZT/TbFe_2$ composite to the variation in H_{dc} at 7 different frequencies of H_{ac} .

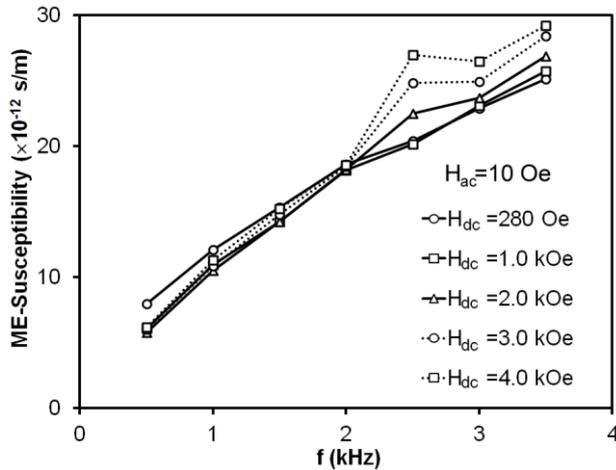


Fig. 5. ME susceptibility of the trilayered $TbFe_2/PZT/TbFe_2$ composite as a function of frequency measured at 5 different H_{dc} .

Due to its unique characteristic, the frequency of 2.5 kHz is chosen along with 500 Hz for the further investigation on the effect of the H_{ac} magnitude. In Fig. 6, the ME response at 3 other H_{ac} values (5, 15 and 20 Oe) resemble that measured using $H_{ac} = 10$ Oe at each frequency and the ME susceptibility at both frequencies is clearly decreased with increasing H_{ac} . Such reduction is originated from the saturation of magnetization rotation. The induced polarization consequently saturates and the ratio in Eq. (1) is diminished by the increase in H_{ac} . In the case of 500 Hz in Fig. 6(a), the peak is lowered with a

slight shift to smaller H_{dc} by the increase in H_{ac} . In Fig. 6(b), the ME susceptibility is rather insensitive to the change of H_{dc} up to 1 kOe. Above 1 kOe, the characteristic linear variation at 2.5 kHz is found at every H_{ac} with the smallest slope in the case of 20 Oe.

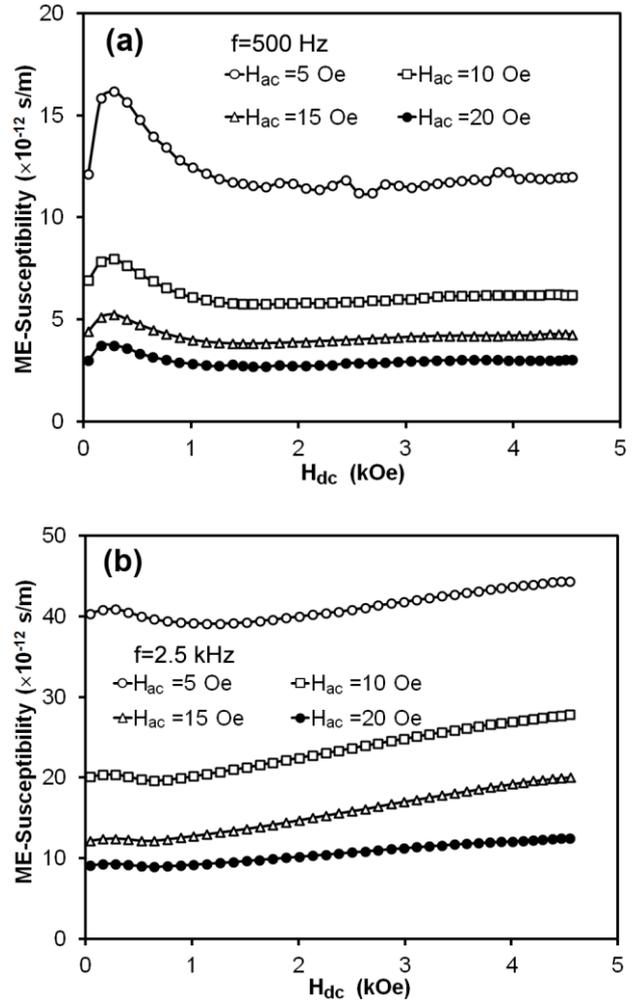


Fig. 6. ME responses of the trilayered $TbFe_2/PZT/TbFe_2$ composite to the variation in H_{dc} at 4 different magnitudes of H_{ac} , measured at (a) 500 Hz and (b) 2.5 kHz.

4. Conclusions

In summary, it is demonstrated that $TbFe_2$ films can be sputtered on both sides of a PZT disk to create a laminate ME composite. The H_{ac} affects both magnitude and characteristic of the ME effect. Due to the saturation in induced polarization, the magnitude of ME susceptibility is dropped by the increase in H_{ac} from 5 to 20 Oe. In a low frequency regime of H_{ac} (< 2.5 kHz), the ME susceptibility is increased with increasing H_{dc} and reaches a maximum at a characteristic bias field of 280 Oe before falling beyond the characteristic bias field. With further increases in H_{dc} , the ME susceptibility is rather constant. In a high frequency regime of H_{ac} (> 2.5 kHz),

the ME susceptibility is steadily increased by the increase in H_{dc} . At 2.5 kHz, the linear ME variation is observed in the H_{dc} range from 800 Oe to 4500 Oe.

Acknowledgments

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