Closed magnetic circuit FeGa/BaTiO$_3$/FeGa sandwich structure for high magnetoelectric effect

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ABSTRACT

Closed magnetic circuit FeGa/BaTiO$_3$/FeGa sandwich structure was fabricated to improve the magnetoelectric effect in a two-phase magnetoelectric system. The magnetoelectric voltages and coefficients were characterized under a static magnetic field in a transverse and an alternating magnetic field in the circumferential. The influences of the field strength are discussed. Calculations predict that the magnetoelectric voltage coefficient peak is located in 67.9 kHz. The measured magnetoelectric voltage coefficient peaks at 68 kHz at 1.88 V/cm Oe. A linear relationship is observed between the magnetoelectric voltage and the alternating field strength under a static field of 340 Oe. The property is attractive for closed magnetic circuit sandwich structure as candidate in current transfer transducer device.

1. Introduction

Recently, magnetoelectric (ME) materials have attracted great interest due to potential applications in multifunctional devices [1,2]. However, the Néel temperature of the single-phase magnetoelectric materials is usually under 140 K [3], only a few single-phase magnetoelectric materials exhibit ME effect and the ME voltage is hard to be detected at room temperature [4], thus they are unable to be used for practical application. In recent years, strong ME coupling has been reported in laminate ME composites of giant magnetostrictive and piezoelectric materials (such as Tb$_{1-x}$Dy$_x$Fe$_2$ (Terfenol-D) and Pb(Mg$_{1/3}$Nb$_{2/3}$)$_3$O$_3$–PbTiO$_3$ (PMN-PT)) [5,6]. Their applications have been realized in multifunctional devices such as electro-magnetic interference, actuators and transducers [7–9]. Though ME coefficient of 7.5 V/cm Oe has been obtained in shear-mode laminated PMN-PT/Terfenol-D composite [10], this composite structure encounters difficulties in practical applications because Terfenol-D is expensive and easily oxidized. In addition, PMN-PT is environmental-unfriendly for its lead content. In this paper, FeGa alloy and BaTiO$_3$ (BTO) oxide are laminated to form closed magnetic circuit FeGa/BaTiO$_3$/FeGa sandwich structure for high magnetoelectricity. FeGa with low hysteresis, high tensile strength, good machinability, and low cost is used as the magnetic phase [11,12]. The piezoelectric BTO, with nothing injurious to the environment [13], is used as the piezoelectric core in the sandwich structure.

To make the ME coefficient $\alpha_{ME}$ measurable, the following expression is considered [14]:

$$\alpha_{ME} = \frac{V}{H_{AC}} \cdot d,$$  

(1)

where $V$ is the voltage generated due to the magnetoelectric effect, $H_{AC}$ is the amplitude of the sinusoidal magnetic field and $d$ is the thickness of the sample or, to be more accurate, the effective distance between the electrodes of the piezoelectric phase. From Eq. (1), $\alpha_{ME}$ can be improved by decreasing $H_{AC}$ at constant $V$. Our previous work used soft magnetic material to decrease $H_{AC}$ [15], and 2.18 mV/cm Oe was achieved in FeCoV film based sandwich structure. As the “magnetic poles” appear at the double end surface when the material is non-infinite or magnetic circuit unclosed, a demagnetizing field ($H_d$) comes into existence in the material being magnetized working in the opposite direction of the applied field. $H_{AC}$ is therefore decreased by the amount of $H_d$. High aspect ratio samples or closed magnetic circuit samples minimize or completely avoid $H_d$. Huang Giang et al. analyzed the enhancement of the ME effects with a narrower width by taking into account of the demagnetization contribution [16]. Leung et al. developed a ring-type electric current sensor for detection of vortex magnetic fields focusing on electric current sensitivity [17]. Dong et al. studied voltage effect in a ring-type magnetoelectric laminate [18]. However, all of the authors did not consider the effect of the demagnetizing and the measurements were only performed under constant field. This paper investigates the effect of the demagnetizing field and the ME response of the closed magnetic circuit FeGa/BTO/FeGa...
sandwich structure with respect to the constant magnetic field, frequency and the alternating field.

2. Experimental details

In making FeGa rod, Fe-19at.% Ga alloy was arc melted several times under argon atmosphere to insure homogeneity. The rod was grown by Bridgman method in axial orientation for good magnetostrictive property [19]. The texture of the rod was examined by X-ray diffraction (XRD) and metallurgical microscope.

Disks of 0.8 mm in thickness were then sliced from the rod, then drilled in the center to achieve “close loop” of outer diameter 28 mm and inner diameter 10 mm. The magnetization and magnetostriction of the hollow disk along the circumference were measured by hysteresis graph of soft magnetic materials [MATS-2010S] and standard strain-gauge technique, respectively.

Commercial BTO discs (φ28 × 2 mm) were also drilled at the center using a diamond bit for a hole of φ10 mm. Table 1 has the basic parameters of the BTO discs from the supplier. The piezoelectric voltage constants $g_{31}$ is 4.4 Vm/N. Two hollow FeGa disks and one hollow BTO disk are thus assembled into a closed magnetic circuit FeGa/BTO/FeGa sandwich structure using conductive epoxy resin, as illustrated in Fig. 1.

The ME effect of the closed magnetic circuit sandwich structure was measured under constant magnetic field strength ($H_{dc}$) in the circumference direction, see Fig. 1. The induced ME voltage was in the thickness direction. As a small alternating magnetic field was superimposed on the static field, the measurement was “dynamic” [20]. The dynamic magnetization was in a circumference direction and the polarization was oriented in transverse. The static magnetic field $H_{dc}$ was controlled by a DC source [GP-HP-L/±65 A] variable from 0 to 800 Oe, and the alternating field $H_{ac}$ variable from 0.1 Oe to 0.25 Oe was generated by a 10 N coils which was supplied by a signal generator [CA1640–02, CALTEK], the strength of which was calculated by Eq. (2) [21].

$$\mathbf{H} = \frac{\mathbf{N} \mathbf{I}}{\mathbf{L}_m}$$

where $\mathbf{N}$ is the number of turns, $\mathbf{I}$ is the current and $\mathbf{L}_m$ is the average magnetic circuit length. The static magnetic field strength $H_{dc}$ was measured by Tesla meter [T-6 TYPE, ISAS]. The electric signal produced by the sample was measured through a lock-in amplifier [LI-5640].

3. Results and discussion

3.1. Texture of the FeGa rod

Fig. 2A plots XRD results of the FeGa rod, confirming the A2 structure with three major peaks (110), (200) and (211). The cross-section along the radial direction (Fig. 2B(a)) indicates equal-axial grains. Along the axial direction (Fig. 2B(b)), only a few grains are seen. These indicate that the FeGa rod obtained is heavily textured along the axial or [110] orientation.

3.2. Magnetization and magnetostriction of the hollow FeGa disk

In the closed magnetic circuit sandwich structure, the compressive stress in the BTO ($\sigma$) is expressed by [22]:

$$\sigma = \frac{2E_f E_t f_t \Delta e_0}{(1 - \nu)(E_t f_t + E_t f_b)}.$$  

where, $E_f$ is the elastic modulus, $t$ is the thickness and $\Delta e_0$ is the linear strain of the FeGa layer, and $\nu$ is the Poisson’s ratio. The subscript $f$ or $b$ represents FeGa or BTO, respectively. From Eq. (3), the stress in the BTO layer enhances with linear strain of the FeGa layer increasing. The voltage output from the piezoelectric layer ($V_i$) in [22]:

$$V_i = 2g_{31} \times t \times \sigma.$$  

Table 1

<table>
<thead>
<tr>
<th>Material</th>
<th>Density ($10^3$ kg/m$^3$)</th>
<th>$g_{31}$ (10$^{-2}$ Vm/N)</th>
<th>$Y_{11}$ (10$^8$ N/m$^2$)</th>
<th>$S_{11}$ (10$^{-12}$ m$^2$/N)</th>
<th>Poisson’s ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaTiO$_3$</td>
<td>5.6</td>
<td>4.4</td>
<td>119</td>
<td>8.4</td>
<td>0.33</td>
</tr>
</tbody>
</table>

Fig. 1. Schematic diagram of magnetoelectric measurement setup and the closed magnetic circuit sandwich structure sample.

Fig. 2. X-ray diffraction (A) and metallurgical microscope result (B) of the FeGa rod in radial (R(a)) and axial (R(b)) directions.

where $g_{31}$ is the piezoelectric voltage constant, $t$ is thickness and $\sigma$ is the stress in the BTO (c.f., Eq. (3)). From Eq. (4), the induced ME voltage from the composite is proportional to the stress of the BTO layer. As such, the ME voltage is proportional to the magnetostriction of FeGa layer. Inserting Eq. (3) into Eq. (4), then into Eq. (1),

$$\alpha_{ME} = \frac{V_i}{t \times H_{ac}} = \frac{2g_{31}}{(1 - \nu)(E_f f_t + E_t f_b)} \times \frac{2E_f f_t \Delta e_0}{H_{ac}}.$$  

Eq. (5) indicates that higher ME coefficient ($\alpha_{ME}$) may be obtained if the ratio of the strain over the magnetic field ($\Delta e_0/H_{ac}$) increased.

When $H_{ac}$ is small, $\Delta e_0$ is less than 0.1 ppm, which is immeasurable by standard strain-gauge. Assuming the magnetostriction is linear, the ratio of saturated magnetostriction over the saturated field is equal to $\Delta e_0/H_{ac}$. Fig. 3 is the magnetization and magnetostriction property of the hollow FeGa disk, examined along circumference. Due to zero demagnetization, only about 45 Oe is needed...
to almost saturate the hollow FeGa disk with saturated magnetostriction of 25 ppm along circumference. In comparison, about 1000 Oe is needed to saturated the FeGa rod and the saturated magnetostriction is about 100 ppm [23]. The orientation of the hollow FeGa disk is perpendicular to the disk surface, which leads to the saturated magnetostricition along the circumference 3/4 less than that of the rod. The saturated field of the FeGa solid disk along thickness direction is larger than FeGa rod due to the demagnetization effect [23]. Thus, the ratio of satuarte magnetostriction over satuarte field for the solid disk is less than that of the rod (100 ppm/1000 Oe [23]). According to Fig. 3, the ratio in the hollow FeGa disk is 25 ppm/45 Oe or 5 folds larger than that of the rod (100 ppm/1000 Oe). From Eq. (5), higher ME coefficient (ME) will be obtained in closed magnetic circuit sandwich structure.

3.3. Resonance frequency of the closed magnetic circuit sandwich structure

\[ f_r = \frac{R}{\pi D} \sqrt{\frac{1}{\rho S_{11}(1 - \nu^2)}} \]  

Here \( D \) is the outer diameter of the loop, \( R \) has a relationship with the ratio of the outer and the inner diameter. In this paper \( R = 1.5 \) is calculated according to Ref. [26], \( \rho \) is the average density, \( \nu \) the Poisson’s ratio. The average equivalent elastic compliance \( S_{11} \) is given by [27]:

\[ S_{11} = V_2 S_{11}^{F_2} + V_1 S_{11}^{F_1} + \frac{V_2 S_{11}^{B_2} + V_1 S_{11}^{B_1}}{V_2 + V_1}, \]  

where \( V_1, V_2 \) and \( V_b \) are the volume fractions of two hollow FeGa plates alone and one hollow BTO plate alone, \( S_{11}^{F_1}, S_{11}^{F_2} \) and \( S_{11}^{B} \) are the elastic compliances of FeGa and BTO. Using the following parameters (data of BTO from Table 1 and FeGa from Datta et al. [19]), \( R = 1.5, D = 28 \text{ mm}, \rho = 6.53 \times 10^3 \text{ kg/m}^3, S_{11}^{F_1} = 16.95 \times 10^{-12} \text{ m}^2/\text{N}, S_{11}^{F_2} = 8.4 \times 10^{-12} \text{ m}^2/\text{N}, \nu = 0.33 \), Eq. (6) yields 67.9 kHz resonance frequency for the closed magnetic circuit sandwich structure. Fig. 4 plots \( \alpha_{ME} \) as a function of \( H_{DC} \) frequency. The results show that the closed magnetic circuit sandwich structure has clear resonance peaks. \( \alpha_{ME} \) peaks at 1.88 V/cm Oe at resonance frequency of \( f_r = 68 \text{ kHz} \). The experimental result of the resonance frequency agrees well with that calculated (67.9 kHz according to Eq.(6)).

3.4. Magnetoelectric effect of the closed magnetic circuit sandwich structure

Fig. 5 shows the influence of the DC magnetic field \( H_{DC} \) on ME voltage coefficient \( \alpha_{ME} \) of the closed magnetic circuit FeGa/BTO/FeGa sandwich structure increases with increasing \( H_{DC} \) and peaks at 1.88 V/cm Oe at \( H_{DC} = 340 \text{ Oe} \) under 68 kHz. When \( H_{DC} > 340 \text{ Oe} \), \( \alpha_{ME} \) decreases with increasing \( H_{DC} \). From phenomenological domain rotation model, magnetostriction is caused by domain rotation, thus 180° rotation results in zero while 90° rotation gives rise to maximum strain [28]. In equilibrium, the domain orientations are completely random. Upon application of a magnetic field, the domains rotate to line up along the field direction.
As the angle between the two fields $H_{AC}$ and $H_{DC}$ is perpendicular, competition between rotation in $H_{DC}$ and $H_{AC}$ direction causes $z_{ME}$ to peak at certain field strength (see Fig. 5(b)).

Fig. 6 exhibits the dependence of induced ME voltages on $H_{AC}$ for the closed magnetic circuit sandwich structure. The induced ME voltages are a linear function of $H_{AC}$. At $H_{AC}$ of 340 Oe and $H_{DC}$ of 0.25 Oe, the ME voltage is about 10 mV at resonance frequency ($f_{r}$ = 68 kHz). Both are eight times higher than that in the solid disk [23]. The $H_{DC}$ of 340 Oe (in comparison, the solid disk needs a $H_{DC}$ of 750 Oe [23]) is easily obtained in practice by permanent magnet and the linear relationship between ME voltage and $H_{AC}$ is a great condition to be used in design alternating current sensors.

4. Conclusions

1. A closed magnetic circuit FeGa/BaTiO$_3$/FeGa sandwich structure avoids demagnetizing field thus improves the magnetoelectric (ME) effect in the two-phase ME system. With the closed magnetic circuit design, the strain versus magnetic strength is five times that obtained in the traditional “solid” counterpart.

2. In the closed magnetic circuit sandwich structure under radial vibration, the Magnetoelectric coefficient $z_{ME}$ peaks at 1.88 V/cm Oe and 68 kHz, agreeing well with the calculated resonance frequency at which the paramount takes place.

3. Magnetoelectric coefficient $z_{ME}$ depends strongly on direct current magnetic field strength ($H_{DC}$). The peak value of $z_{ME}$ occurs at $H_{DC}$ = 340 Oe, which is easily attainable in practice. With the closed magnetic circuit design the magnetoelectric voltage varies linearly with the alternative current magnetic field strength ($H_{AC}$). As the demagnetizing field is avoided, the magnetoelectric voltage is eight times more sensitive with $H_{AC}$ than that in the solid counterpart.

4. As the FeGa/BaTiO$_3$/FeGa system contains nothing injurious to the environment, this system further adds to benefit applications in high voltage and current transmission, among others.

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