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Enhanced Giant Magnetoelectric Effect in Laminate Composites of FeCuNbSiB/FeNi/PZT

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A novel laminate composite of FeCuNbSiB/FeNi/PZT is proposed, where FeCuNbSiB has a permeability of around 100000, which is much larger than that of FeNi. The high-permeability FeCuNbSiB was laminated with piezomagnetic FeNi rather than attached to its ends. It is expected that the effect produced by the high permeability will act on the whole of the piezomagnetic layer. While a FeNi layer was laminated with a FeCuNbSiB layer, the strong demagnetization produced by the latter was expected to be imposed on the FeNi layer as well as the applied fields. The distribution of applied fields was altered by the high-permeability material (both bias and ac field) and the field variation positively contributed to the ME effect in piezomagnetic/piezoelectric composites. Thus the ME voltage coefficient along with the field sensitivity were improved.

Keywords : piezomagnetic/piezoelectric composite, magnetoelectric effect, permeability

1. Introduction

The laminate composites of piezomagnetic and piezoelectric materials exhibit a giant magnetoelectric (ME) effect at room temperature and the effect originates from the magneto-mechano-electric coupling between the two materials [1]. Based on this, it is naturally accepted that the magnetostrictive (in piezomagnetic phase) and mechanoelectric (in piezoelectric phase) properties and their interactions are critical for the ME performances. To obtain better giant ME effects, most efforts have been focused on investigating the two properties and to design composite architectures enhancing the interactions [2]. Dong investigated the incorporation of high permeability material in magnetostrictive/piezoelectric laminate [3, 4]. By attaching the high permeability bars to either ends of the laminate, the flux is concentrated and the ME voltage coefficient at low biases is enhanced.

Several analytical models have been established to predict the ME effects of piezomagnetic and piezoelectric laminate composites [5, 6]. Among the models, fewer are connected with permeability. Dong *et al.* employed equivalent circuit approaches to model ME behavior of piezomagnetic/piezoelectric composites [7]. This model has been extensively used in laminate configuration design and ME response analysis. In the equivalent circuit, the effect of the piezomagnetic phase is equivalent to a voltage source, the outcome of the ME effect is the electric outputs. This model well predicts the optimal geometric ratio related to the layer thicknesses that maximizes the ME voltage. Based on the equivalent model, Yang *et al.* introduced a resistor in the ME equivalent circuit, which represents the magnetic, mechanical, and dielectric losses in the magnetic-mechanical-electric transduction process [8]. Thus the ME output given by the equivalent circuit is becoming closer to the experimentals. It was also illustrated that among these losses, the discontinuity of stress coupling at interfaces of a heterostructure is overwhelming and the other losses can be ignored. From the ME equivalent circuit, Yang *et al.* derived the ME voltage sensitivity to the bias (dc fields) by letting the piezomagnetic constant be a function of dc fields [9]. The ME equivalent circuit is commonly used. However, the effect of permeability in the piezomagnetic phase is not presented in the equivalent circuit. Thus the equivalent circuit is unable to account for the ME effect of composites incorporated with high-permeability materials.

Conceptually, it is not adequate to describe ME performances while being ignorant of permeability. In the

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Bichurins' ME voltage expression, the longitudinal ME voltage coefficient is inversely proportional to $\mu_0[1-(\mu_{33}-1)\nu]$, where μ_{33} is the relative permeability of the piezomagnetic phase and ν is the volume ratio of the piezoelectric phase in the composite volume. And the transverse ME voltage coefficient expression shows no relation to the magnetic permeability [10].

Laletsin *et al.* fabricated trilayer structures of ferromagnetic iron, nickel, or cobalt as well as piezoelectric PZT and the thickness of the metals and PZT was 0.4 mm [11]. For MEVCs, a maximum value of around 90 V/cm *Oe* at resonance was measured for both Co-PZT-Co and Ni-PZT-Ni, and a maximum value of 30 V/cm *Oe* was measured for Fe-PZT-Fe. This clearly demonstrated that the permeability is critical to the ME effect in Piezomagnetic/Piezoelectric Composites and somewhat conflicted with Bichurins' ME voltage expression.

Inspired by these facts, we developed new ME composites by laminating a high-permeability FeCuNbSiB layer with a FeNi/PZT composite based on the analysis of field variation induced by the magnetic layers in piezomagnetic/piezoelectric composites. The ME effect of the new heterostructures was eminently improved. The optimal bias field for FeCuNbSiB/FeNi/PZT was largely reduced at the same time as the ME voltage coefficient and the field sensitivity were enhanced.

2. The Field Variation Induced by FeNi and FeCuNbSiB

In all the current analysis regarding the ME effect in piezomagnetic/piezoelectric composites, the magnetic fields acting on heterostructures are assumed to be uniform. However, as the piezomagnetic phase in ME composites is magnetic, in magnetic fields its magnetic permeability will effect and produce a flux concentration. This in turn alters the field distribution around and inside a composite even when a field is uniformly applied. Due to the flux concentration, the field can no longer be taken as being uniformly distributed and evenly acting on an ME composite.

Fig. 1(a) gives the flux concentration produced by a FeNi plate (12 mm × 6 mm) in a uniform dc field, which is simulated using Ansoft Maxwell. It demonstrates that the uniformity is altered by the FeNi plate. Fig. 1(b) illustrates the field along the longitudinal direction and Fig. 1(c) depicts the derivative of the longitudinal field with respect to the coordinate. It is obvious that the average field inside the plate is reduced and the uniform distribution is altered and sharp variations at either end occur. Fig. 2 shows the flux concentration from the FeCuNbSiB/FeNi laminate. The concentration at either end is denser than that at the ends of a FeNi plate. This suggests that the field variation is even sharper at either end of the FeCuNbSiB/FeNi laminate.

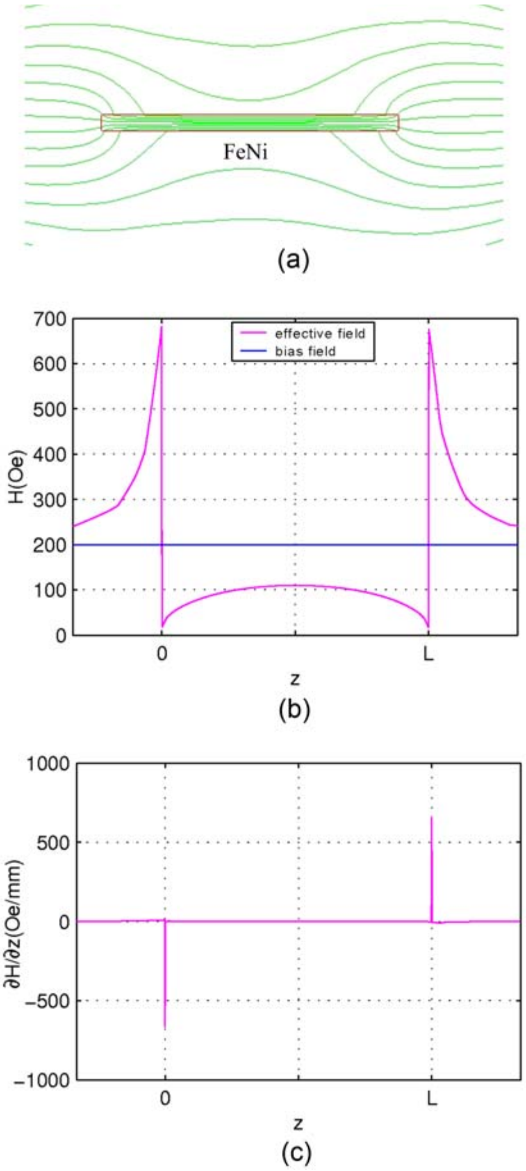


Fig. 1. (Color online) Variation of a uniformly applied field caused by a FeNi plate (a) Flux concentration caused by magnetic material (b) Field distribution along longitudinal direction (c) The derivative with respect to the longitudinal coordinate.

3. The ME Voltage Coefficient in Consideration of the Field Variation Induced by Magnetic Layers

In the proposed heterostructures, the magnetic layer is longitudinally magnetized parallel to the motion direction

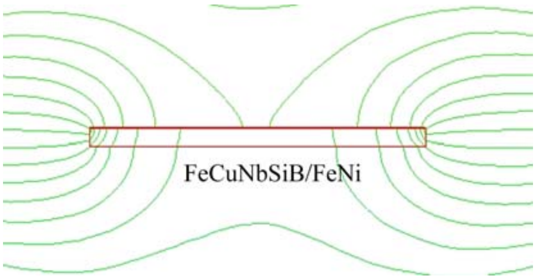


Fig. 2. (Color online) Flux concentration caused by FeCuNbSiB/FeNi laminate.

and the piezoelectric layer is polarized perpendicular to the motion in the thickness direction. Designating axis z as the strain direction of a composite, the movement of the composite is described from Newton's second law as:

$$\bar{\rho}\frac{\partial^2 u}{\partial t^2}=\frac{n}{s_{11}^H}\left(\frac{\partial^2 u}{\partial z^2}-d_{11,m}\frac{\partial H}{\partial z}\right)+\frac{1-n}{s_{11}^E}\left(\frac{\partial^2 u}{\partial z^2}-d_{31,p}\frac{\partial E}{\partial z}\right) \tag{1}$$

where u is the displacement, H is the magnetic field, E is the electric field; s_{11}^E , $d_{31,p}$ and ε_{33}^T are the elastic compliance at constant E, the piezoelectric constant, and dielectric constant, respectively; $\bar{\rho}=\rho_p A_p+\rho_m A_m/A_p+A_m$, $n=A_m/A_p+A_m$, ρ_p, and ρ_m are the mass densities of the piezoelectric and magnetic layers, and A_p, A_m are the cross-sectional areas. Here the magnetically induced strain in the piezomagnetic layer is assumed to be fully transferred to the piezoelectric layer. If the strain loss caused by bonding is not ignorable [12], it can be described by a coupling factor as indicated in [10]. E is not related to z, thus $\partial E/\partial z=0$. However, even if the applied field is uniform due to the demagnetization from the permeability of magnetic layer(s), the effective field becomes nonuniformly distributed and especially sharply varied at either and of an ME composite. H should be the effective field component which is the synthesis of the applied and the demagnetized fields. When the applied field is uniform, sharp variations of the effective field symmetrically occur at the ends of the composite. Given that the length of the composite is L, in general

$$\left.\frac{\partial H}{\partial z}\right|_{z=0}=-\left.\frac{\partial H}{\partial z}\right|_{z=L} \tag{2}$$

For simplification, the derivative of the field with respect to z is approximated as

$$\frac{\partial H}{\partial z}\approx\begin{cases}\left.\dfrac{\partial H}{\partial z}\right|_{z=0} & z=0\\ -\left.\dfrac{\partial H}{\partial z}\right|_{z=L} & z=L\\ 0 & otherwise\end{cases} \tag{3}$$

The field variation actually functions by producing forces that result in a strain at either end face of the ME laminates. The strain is

$$S_\xi=(1+\cos KL)\frac{\xi_0}{K\bar{\mathrm{v}}^2} \tag{4}$$

where

$$\xi_0=\left.\frac{\partial H}{\partial z}\right|_{z=0}\cdot d_{11,m}\cdot\frac{n}{s_{11}^H}/\bar{\rho},\ \bar{\mathrm{v}}^2=\frac{\frac{n}{s_{11}^H}+\frac{1-n}{s_{11}^E}}{\bar{\rho}},\quad K=\omega^2/\bar{\mathrm{v}}^2\ .$$

ω is the angular frequency of the magnetic drive.

Under free-free boundary conditions, the final output voltage is

$$V=\frac{\varphi_m\varphi_p H(0)+\varphi_p F_\xi}{(Z_2+Z_1/2)\cdot j\omega C_0+\varphi_p^2} \tag{5}$$

where $F_\xi=-(A_m+A_p)/\bar{s}\cdot S_\xi$; φ_m, φ_p, Z_1, Z_2 and C_0 are parameters in the ME equivalent circuit. This demonstrates that the field variation induced by the magnetic layers positively contribute to the ME effect. The sharper the variation is, the more enhanced the ME effect becomes. From the analysis, it is predicted that at a given piezomagnetic coefficient, a higher permeability will produce a stronger ME effect.

4. Fabrication and Experiments

From the analysis regarding the laminated composites, it is known that the ME response is connected with the thickness ratio of magnetostrictive and piezoelectric layers. And the output voltage is proportional to the thickness of the piezoelectric layer. For easy fabrication, prototypes of the proposed 3-phase laminate composite were fabricated with FeCuNbSiB (12 mm × 6.5 mm × 0.028 mm), FeNi (12 mm × 6 mm × 0.6 mm), and PZT (12 mm × 6 mm × 0.8 mm) as schematically shown in Fig. 3. FeNi functioned as a piezomagnetic layer and its relative permeability is around 2.5 × 10^3. The soft magnetic FeCuNbSiB was used to enhance the permeability of the magnetic phase in the composite as its relative permeability is much higher than that of FeNi at about 5 × 10^5. The FeNi layer was longitudinally magnetized and the PZT layer was polarized in the thickness direction. The in-plane dimensions of the FeCuNbSiB, FeNi, and PZT

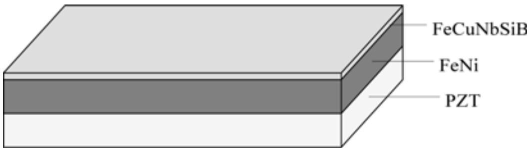


Fig. 3. FeCuNbSiB/ FeNi/PZT laminate composite.

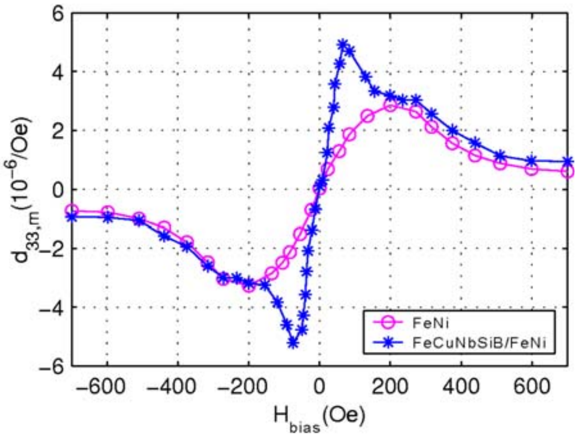
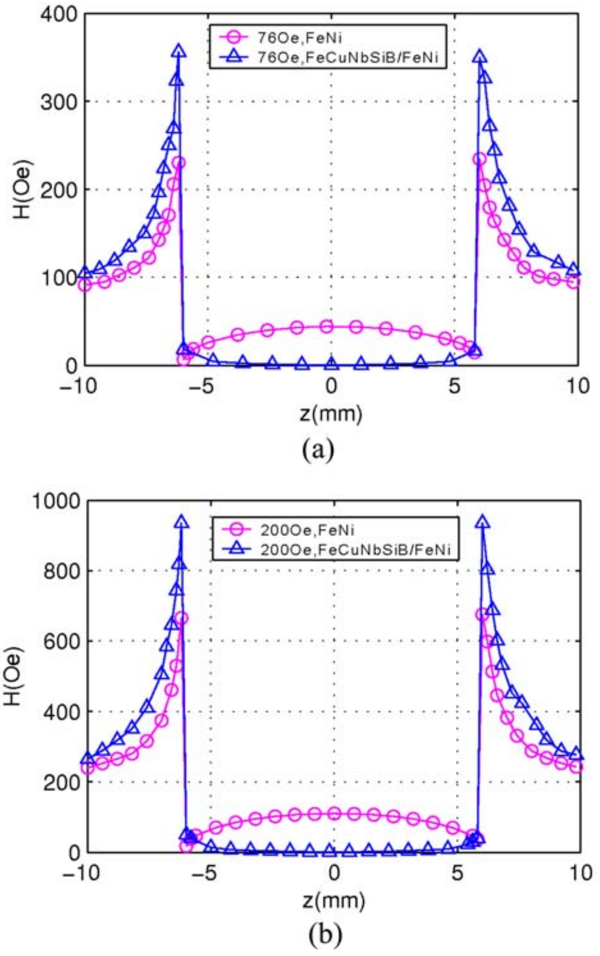


Fig. 4. (Color online) Measured effective dynamic coefficients vs. bias fields.

layers were the same at 12 m × 8 mm and their thicknesses were respectively 0.028 mm, 0.6 mm, and 0.8 mm.

The effective piezomagnetic coefficients of the FeNi plate and FeCuNbSiB/FeNi laminate were measured with respect to the bias field, as shown in Fig. 4. The image illustrates that the effective piezomagnetic coefficient of FeNi plate is nonlinearly related to the bias field. The effective piezomagnetic coefficient of the FeCuNbSiB/FeNi laminate was greatly enhanced until the FeNi piezomagnetic coefficient retained its maximum. The maximum of the FeCuNbSiB/FeNi piezomagnetic coefficient was much higher than that of FeNi. FeCuNbSiB has little piezomagnetism. While bonded with a piezomagnetic plate, FeCuNbSiB layer is a mechanical load in magneto-mechanical transduction and will not directly contribute to piezomagnetism. From the above analysis, it is known that part of the magnetically induced strain is attributed to the variation of field distribution at the ends of the piezomagnetic layer, which resulted from the flux concentration produced by the magnetic material in the magnetic field. As shown in Fig. 5(a) and (b), the field variation caused by the FeCuNbSiB/FeNi laminate is larger than that produced by the FeNi plate. Hence with identical piezomagnetic coefficients, the actual piezomagnetically induced strain in the FeCuNbSiB/FeNi laminate is larger than that in the FeNi plate. This accounts for the effective piezomagnetic coefficient growing.

The MEVCs of FeNi/PZT and FeCuNbSiB/FeNi/PZT composites were measured under various bias fields as show in Fig. 6. The image demonstrates that: 1) the optimal bias for the FeNi/PZT composite is 200 *Oe* and the optimal bias for the FeCuNbSiB/FeNi/PZT composite is less at around 76 *Oe*; 2) the MEVCs of FeCuNbSiB/FeNi/PZT composite are larger than that of the FeNi/PZT composite and the maximum value for the FeCuNbSiB/FeNi/PZT composite is more than 80% higher than the



(a)

(b)

Fig. 5. (Color online) Field distributions inside and outside a FeNi plate and FeCuNbSiB/FeNi laminate (a) H_{dc} = 76 *Oe* (b) H_{dc} = 200 *Oe*.

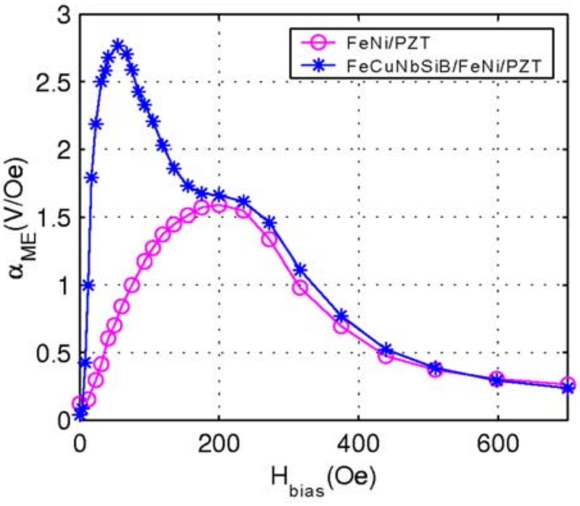


Fig. 6. (Color online) Measured ME voltage coefficients vs. bias fields.

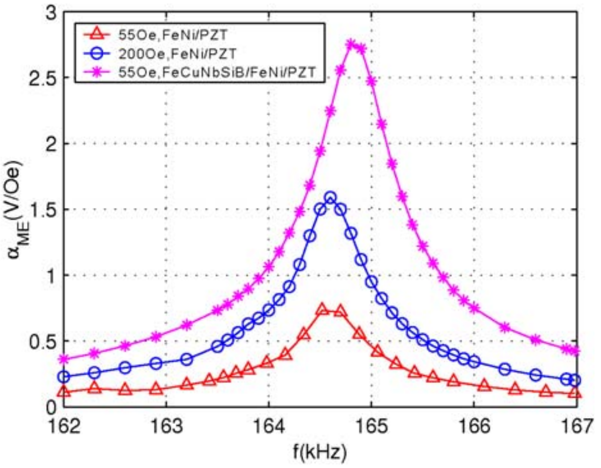


Fig. 7. (Color online) Measured ME voltage coefficients vs. frequency under various biases.

maximum value for the FeNi/PZT composite. The results show that due to the higher permeability of FeCuNbSiB, a stronger flux concentration occurs with FeCuNbSiB/FeNi, this in turn reduces the optimal field bias for the FeCuNbSiB/FeNi/PZT composite. At the same time, the stronger flux concentration produces sharper variation of the field distribution near the end faces of the FeCuNbSiB/FeNi/PZT composite. Consequently, a higher MEVC is achieved with the given piezomagnetic property of FeNi. Fig. 7 gives the comparison of ME voltage coefficients of FeNi/PZT and FeCuNbSiB/FeNi/PZT vs. frequency. From Figs. 6 and 7, it is seen that the bias field will influence the ME voltage coefficients.

5. Conclusion

In summary, the proposed ME composite exhibited better performances with respect to the ME effect and it is applicable in transducer or sensor applications. The measured ME performances of the fabricated prototypes indicate that the optimal bias field for the proposed composite obviously decreases from around 200 *Oe* for the two-phase composite to 55 *Oe*. At the same time, the ME voltage coefficient greatly increases from ~1.5V/*Oe* to ~2.8V/*Oe*.

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References

[1] J. Ryu, A. V. Carazo, K. Uchino, and H. Kim, Jpn. J. Appl. Phys. **40**, 4948 (2001).
[2] P. Li, Y. Wen, and L. Bian, Appl. Phys. Lett. **90**, 022503 (2007).
[3] S. X. Dong, J. Zhai, J. Li, D. Viehland, and D. Viehland, J. Appl. Phys. **100**, 124108 (2006).
[4] S. X. Dong, J. Zhai, J. Li, and D. Viehland, Appl. Phys. Lett. **89**, 122903 (2006).
[5] G. Harshe, J. O. Dougherty, and R. E. Newnham, Int. J. Appl. Electromag. Mater. **4**, 145 (1993).
[6] M. Avellaneda and G. Harshe, J. Intell. Mater. Syst. Struct. **5**, 501 (1994).
[7] S. Dong, J. F. Li, and D. Viehland, IEEE Trans. Ultrason. Ferroelectr. Freq. Control. **50**, 1253 (2003).
[8] F. Yang, Y. Wen, P. Li, M. Zheng, and L. Bian, Sens. Actuators A-Phys. **141**, 129 (2008).
[9] C. Yang, Y. Wen, P. Li, and L. Bian, Acta Phys. Sin. **57**, 7292 (2008) (in Chinese).
[10] M. I. Bichurin, V. M. Petrov, and G. Srinivasana, J. Appl. Phys. **92**, 7681 (2002).
[11] U. Laletrin, N. Paddubnaya, G. Srinivasan, and M. I. Bichurin, Magnetoelectric Interaction Phenomena in Crystals-NATO Science Series II, **164**, Eds. M. Fiebig, V. V. Eremenko, and I. E. Chupis, Kluwer Academic Publishers, London (2004) pp.57-64.
[12] W. Yang, Y. Wen, and P. Li, Acta Phys. Sin. **57**, 4545 (2008) (in Chinese).