AIP | Applied Physics Letters

Giant magnetoelectric effect (under a dc magnetic bias of 2 Oe) in laminate composites of FeBSiC alloy ribbons and $Pb(Zn_{1/3},Nb_{2/3})O_3-7\%PbTiO_3$ fibers

Shuxiang Dong, Junyi Zhai, Zhengping Xing, Jiefang Li, and D. Viehland

Citation: Applied Physics Letters **91**, 022915 (2007); doi: 10.1063/1.2757146
View online: http://dx.doi.org/10.1063/1.2757146
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/91/2?ver=pdfcov
Published by the AIP Publishing

APPLIED PHYSICS LETTERS **91**, 022915 (2007)

Giant magnetoelectric effect (under a dc magnetic bias of 2 Oe) in laminate composites of FeBSiC alloy ribbons and $Pb(Zn_{1/3},Nb_{2/3})O_3-7\%PbTiO_3$ fibers

Shuxiang Dong,[a] Junyi Zhai, Zhengping Xing, Jiefang Li, and D. Viehland
Materials Science and Engineering, Virginia Tech, Blacksburg, Virginia 24061

(Received 11 May 2007; accepted 19 June 2007; published online 12 July 2007)

Giant magnetoelectric (ME) voltage and charge coefficients have been found in long-type composites of high-permeability magnetostrictive FeBSiC alloy ribbons laminated together with piezoelectric $Pb(Zn_{1/3},Nb_{2/3})O_3-7\%PbTiO_3$ single crystal fibers. The maximum ME voltage and charge coefficients at low frequencies were 10.5 V/cm Oe and 1 nC/Oe under a notably low dc magnetic bias of 2 Oe; at resonance, these coefficients were dramatically increased to 400 V/cm Oe and 42 nC/Oe, respectively. These values are much higher, and the required dc magnetic bias much lower, than those of previously reported Terfenol-D based ME laminates. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2757146]

The magnetoelectric (ME) effect is an electric polarization response to an applied magnetic field, or conversely a magnetization response to an applied electric field. Only small ME effects are known in single phase materials.[1] Orders of magnitude larger ME coefficients have been reported in two phase composites: in particular, the largest ME coefficients are found in laminated composites of magnetostrictive and piezoelectric layers.[2–9] Large magnetoelectric coefficients offer potential device applications as highly sensitive magnetic field sensors, microwave filters, transformers, and gyrators.[10,11]

Recently,[12] we reported that a three-layer laminate with a (2-1) phase connectivity consisting of high-permeability magnetostrictive FeBSiC alloy layers longitudinally (L) magnetized and piezoelectric $Pb(Ti,Ti)O_3$ (PZT) fibers longitudinally (L) poled had ME voltage coefficients of 22 V/cm Oe at low frequency and of 500 V/cm Oe at resonance. These values are nearly an order of magnitude higher than prior reports for (L-L) Terfenol-D/PZT laminates,[2–9] which up to that time had the highest known values. Here, we report a long-type ME laminate configuration consisting of $Pb(Zn_{1/3},Nb_{2/3})O_3-7\%PbTiO_3$ (PZNPT) single crystal fibers laminated between two long FeBSiC alloy ribbon layers, operated in a longitudinally (L) magnetized and transversely (T) poled (or L-T) mode. Among all known ME composites that are operated in L-T modes, we will see that this FeBSiC/PZN-PT ME configuration has (i) the lowest required dc magnetic bias H_{dc} and (ii) the highest ME voltage and charge coefficients.

Magnetostrictive/piezoelectric laminates operated in L-T mode have been analyzed using an equivalent method.[9] The ME voltage coefficient V_{ME}^{LT} can be given as

$$V_{ME}^{LT} = \left| \frac{dE_1}{dH_3} \right| = \frac{n d_{33,m} g_{31,p}}{n s_{11}^E (1 - k_{31,p}^2) + (1 - n) s_{33}^H}, \tag{1}$$

where n is the magnetic phase thickness ratio, $d_{31,p}$ and $g_{31,p}$ are the longitudinal piezomagnetic and transverse piezoelectric voltage coefficients, s_{11}^E and s_{33}^H are the elastic compliances of the piezoelectric and magnetostrictive layers, and $k_{31,p}$ is the electromechanical coupling coefficient of the piezoelectric layer. Following Eq. (1), the ME charge coupling coefficient for the L-T mode Q_{ME}^{LT} is

$$Q_{ME}^{LT} = \frac{dQ_1}{dH_3} = \frac{n d_{33,m} d_{31,p} A_p}{n s_{11}^E (1 - k_{31,p}^2) + (1 - n) s_{33}^H}, \tag{2}$$

where $d_{31,p}$ is the transverse piezoelectric charge coefficient and A_p is the electrode area of the piezoelectric fiber layer. Equations (1) and (2) show that a high $d_{33,m}$ in the magnetic phase and a high $g_{31,p}$ or $d_{31,p}$ in the piezoelectric one will result in a high V_{ME}^{LT} or Q_{ME}^{LT}. In Table I, some important parameters for Metglas and PZNPT are given. The maximum $d_{33,m}$ of FeBSiC is three to four times larger than that of Terfenol-D, and $d_{31,p}$ of PZNPT crystals is approximately five times larger than that of PZT. Accordingly, much higher values of V_{ME}^{LT} and Q_{ME}^{LT} can be expected for (L-T) FeBSiC/PZNPT-fiber laminates than for Terfenol-D/PZT ones. Furthermore, the extremely high permeability ($\mu_r = 40\,000$) of FeBSiC alloys will dramatically reduce the saturation field, relative to that of Terfenol-D ($\mu_r = 10$), which subsequently will result in a dramatic reduction in the required dc magnetic bias H_{dc}. Furthermore, according to Eq. (2), a ME laminate containing more piezoelectric fibers will have a larger A_p, resulting in a higher ME charge coefficient Q_{ME}^{LT}.

In our experiments, we chose high-permeability FeBSiC alloy ribbons (Metglas Inc, Conway, SC) as the magnetostrictive layers, and PZNPT single crystal fibers poled in a d_{31} configuration (Microfine Materials Tech, Singapore) as the piezoelectric one. A thin PZN-PT crystal plate oriented along [100] in its length direction was cut into fibers of sizes 15 mm in length, 0.4 mm in width, and 0.1 mm in thickness. A piezoelectric fiber layer has a high capacitance in its thickness direction, which is quite thin, therefore, it has potential for achieving high ME charge coupling under a very low dc magnetic bias. The FeBSiC ribbons were 100 mm long, 5 mm wide, and 25 μm thick. Two FeBSiC alloy ribbons and one PZN-PT single crystal fiber layer were then bonded together using an epoxy resin and heated at 80 °C for 30 min, forming a long-type ME composite with a two-dimensionally connected magnetic phase and one-dimensionally connected piezoelectric phase, i.e., a (2-1)

---

[a] Electronic mail: sdong@mse.vt.edu

0003-6951/2007/91(2)/022915/3/$23.00

TABLE I. Electromechanical and magnetoelastic material parameters for FeBSiC and ⟨001⟩-oriented PZN-PT crystals.

	$d_{33,m}$ or $d_{33,p}$	$d_{31,m}$ or $d_{31,p}$	g_{31}	s_{11}^H or s_{11}^E	s_{33}^H or s_{33}^E	k_{33}	k_{31}	$\varepsilon_{33\ \mathrm{eff}}^{T}$ [a]
FeBSiC[b]	1.2×10^{-8} (Wb/N)	-5.8×10^{-9} (Wb/N)		125×10^{-12} (m^2/N)	40×10^{-12} (m^2/N)	0.7		
⟨001⟩ PZN-PT[c]		−2800 (pC/N)	44×10^{-3} (mV/N)	69.0×10^{-12} (m^2/N)	119×10^{-12} (m^2/N)	0.94	0.93	5500

[a] Measured value.
[b] From Metglas Inc, Conway, SC.
[c] From Microfine Materials Tech, Singapore.

phase connectivity. Figure 1 illustrates the structure of our FeBSiC/PZNPT-fiber laminate, in which the FeBSiC layer(s) was (were) longitudinally magnetized and the piezoelectric one transversely poled.

During ME measurements, dc (H_{dc}) and ac (H_{ac}) magnetic fields were applied along the length of the laminates. An electromagnet was used to provide H_{dc}, and a Helmholtz coil was used to generate $H_{ac}=1$ Oe. A lock-in amplifier (SR850) generated a controllable input current to the Helmholtz coil, and was also subsequently used to measure the output voltage induced across the PZN-PT layer of the ME laminate by H_{ac}.

First, we measured the effect of length of the FeBSiC layer on the required H_{dc} for the ME laminate in Fig. 1. Previously,[12] the flux in ME laminates was found to be near linearly increased as the length of the Metglas layer was increased: this is simply because long-type high-μ layers can collect more magnetic flux. In addition, long magnetic layers in ME laminates are important in lowering the required H_{dc}, because it can effectively decrease the demagnetization factor N and reduce the saturation field. Figure 2 shows the required H_{dc} as a function of the length ratio of the Metglas layer to that of the piezoelectric fiber one. The results clearly show that longer Metglas layers result in lower required values of H_{dc}, that is until the magnetic phase approaches saturation.

Next, we measured the ME responses of the FeBSiC/PZN-PT L-T mode laminate of Fig. 1. Figure 3(a) shows the low frequency ($f=1$ kHz) ME voltage coefficient as a function of dc magnetic bias H_{dc}. The maximum value of this coefficient at subresonant frequencies can be seen to be $V_{ME}^{LT}>10$ V/cm Oe under a very low $H_{dc}=2$ Oe: this is approximately five times higher than the highest value for a L-T configuration previously reported, which was for Terfenol-D/PMN-PT ($V_{ME}^{LT}\approx2$ V/cm Oe) (Ref. 9) under a $H_{dc}=500$ Oe. The required $H_{dc}=2$ Oe is dramatically lower than that in previous reports. We then determined that the corresponding maximum value in the ME charge coefficient was $Q_{ME}^{LT}\approx1$ nC/Oe. Figure 3(b) shows V_{ME}^{LT} of the L-T FeBSiC/PZNPT-fiber laminate as a function of ac magnetic drive frequency at $H_{dc}=2$ Oe. A strong resonance enhancement in V_{ME}^{LT} to 400 V/cm Oe can be seen in this figure at a resonance frequency of $f_1=20$ kHz that corresponds to the first-order longitudinal electromechanical resonance. The corresponding value of Q_{ME}^{LT} was determined to be 42 nC/Oe. The inset in Fig. 3(b) shows the impedance/phase-frequency spectrum of the laminate around $f=20$ kHz, which may help to estimate the magnitude of effective electromechanical coupling coefficient k_{31} and piezoelectric coefficient d_{31} in the piezoelectric phase of the laminate. This ME laminate also exhibits two small resonances at $f_2=58$ kHz and $f_3=92$ kHz that correspond to second- and third-order longitudinal modes. Such large ME voltage and charge couplings at resonance could be of importance in power device applications.

Following Eqs. (1) and (2), these large values of the ME coefficients can be attributed to (i) the high apparent piezomagnetic coefficient $d_{33,m}$ of the magnetostrictive FeBSiC alloy phase due to flux concentration, (ii) the high $g_{31,p}$ and $d_{31,p}$ of the PZN-PT single crystal fibers, and (iii) the (2-1) connectivity of the ME laminate configuration that causes a more intimate contact between the two phases. The low value of H_{dc} can be attributed to (i) the high μ of the FeBSiC magnetic layers, (ii) long-type configuration of the ME laminate, and (iii) the large compliance of the thin-type piezoelectric fibers. Finally, we also measured the magnetic field sensitivity of L-T FeBSiC/PZNPT-fiber laminates at $f=1$ Hz and 20 kHz. The experimental data (not given)

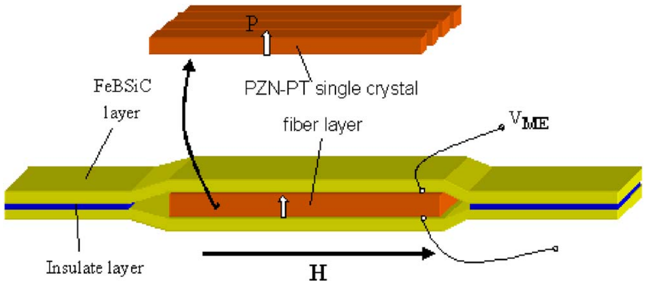

FIG. 1. (Color online) Structure of FeBSiC/PZNPT-fiber laminate.

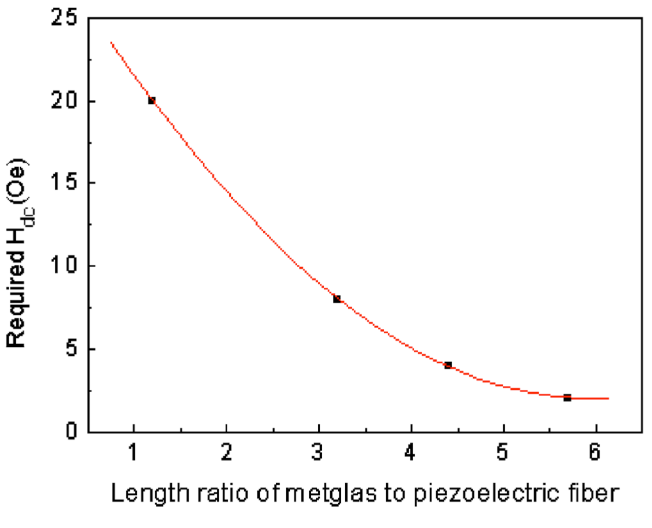

FIG. 2. (Color online) Required H_{dc} as a function of the length ratio of the FeBSiC layer to that of the piezoelectric fiber one.

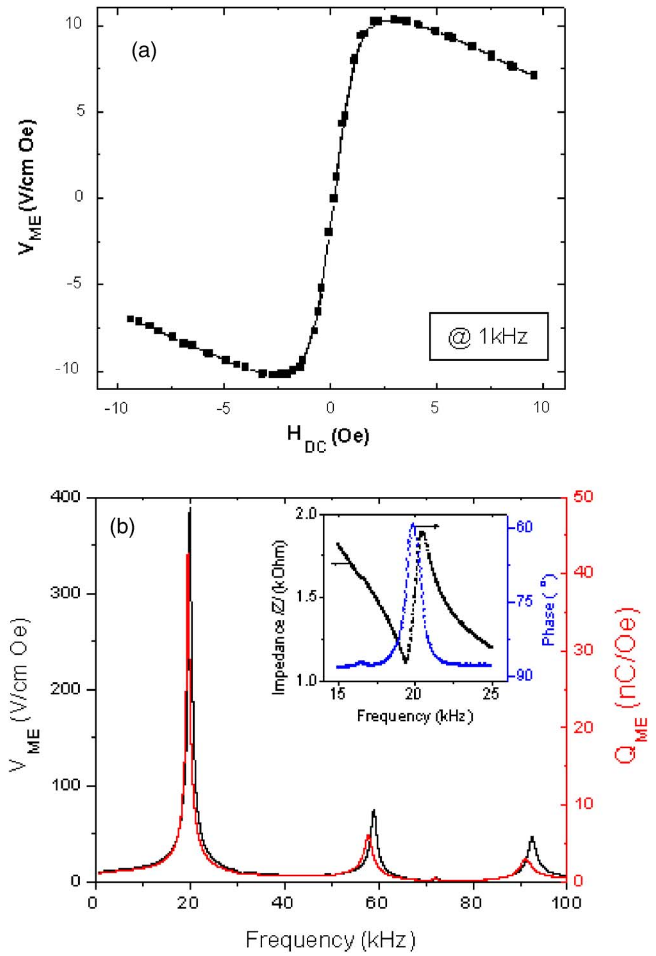

FIG. 3. (Color online) ME voltage coefficients of FeBSiC/PZNPT-fiber laminate: (a) $V_{\mathrm{ME}}^{\mathrm{LT}}$ as a function of dc magnetic bias H_{dc} at $f=1$ kHz and (b) $V_{\mathrm{ME}}^{\mathrm{LT}}$ as a function of ac magnetic drive frequency. The inset in shows the impedance spectrum of the laminate.

showed that this type of ME laminate had a ME sensitivity of approximately 10^{-11} and 10^{-12} T at 1 Hz and resonance, respectively.

In summary, we have found that long-type FeBSiC/PZNPT-fiber laminates operated in L-T mode have large ME voltage and charge coefficients of 10.5 V/cm Oe and 1 nC/Oe, respectively, at low frequency and under a lowest dc magnetic bias $H_{\mathrm{dc}}=2$ Oe, which were dramatically enhanced under resonance drive to 400 V/cm Oe and 42 nC/Oe, respectively. These values of the ME coefficients are five times than that previously reported for any L-T laminates.

The authors gratefully acknowledge the full support by the Office of Naval Research.

[1] V. J. Folen, G. T. Rado, and E. W. Stalder, Phys. Rev. Lett. **6**, 607 (1961).
[2] C.-W. Nan, Phys. Rev. B **50**, 6082 (1994).
[3] Z. Shi, C. W. Nan, J. Zhang, N. Cai, and J.-F. Li, Appl. Phys. Lett. **87**, 012503 (2005).
[4] S. X. Dong, J. F. Li, and D. Viehland, Appl. Phys. Lett. **85**, 5305 (2004); J. Appl. Phys. **95**, 2625 (2004).
[5] G. Srinivasan, E. T. Rasmussen, and R. Hayes, Phys. Rev. B **67**, 014418 (2003).
[6] S. X. Dong, J. Zhai, F. Bai, J. Li, D. Viehland, and T. Lograsso, J. Appl. Phys. **97**, 103902 (2005).
[7] G. Srinivasan, E. Rasmussen, J. Gallegos, R. Srinivasan, Y. Bokhan, and V. Laletin, Phys. Rev. B **64**, 214408 (2001).
[8] M. Bichurin, V. Petrov, and G. Srinivasan, Phys. Rev. B **68**, 054402 (2003).
[9] S. X. Dong, J. F. Li, and D. Viehland, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **50**, 1253 (2003); **51**, 794 (2004).
[10] S. X. Dong, J. F. Li, and D. Viehland, Appl. Phys. Lett. **85**, 2307 (2004); **83**, 2265 (2003); **85**, 3534 (2004); **89**, 243512 (2006); **84**, 4188 (2004).
[11] Y. K. Fetisov and G. Srinivasan, Appl. Phys. Lett. **88**, 143503 (2006).
[12] S. X. Dong, J. Y. Zhai, J. F. Li, and D. Viehland, Appl. Phys. Lett. **89**, 252904 (2006).