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Citation: Applied Physics Letters **89**, 252904 (2006); doi: 10.1063/1.2420772
View online: http://dx.doi.org/10.1063/1.2420772
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/89/25?ver=pdfcov
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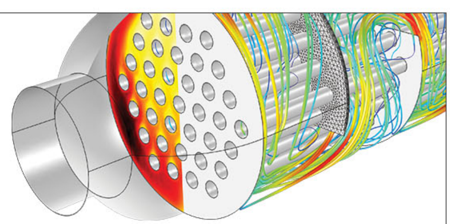

APPLIED PHYSICS LETTERS **89**, 252904 (2006)

Near-ideal magnetoelectricity in high-permeability magnetostrictive/piezofiber laminates with a (2-1) connectivity

Shuxiang Dong,[a)] Junyi Zhai, Jiefang Li, and D. Viehland
Materials Science and Engineering, Virginia Tech, Blacksburg, Virginia 24060

(Received 2 October 2006; accepted 14 November 2006; published online 20 December 2006)

Theoretically, the two-phase laminated configurations should have even much higher magnetoelectric (ME) effects—however, prior experimental studies have failed to find such an enhancement. Here, the authors report the unleashing of the potential of the (2-1) connectivity configuration: a piezofiber (one-dimension connectivity) layer laminated between two high-permeability magnetostrictive FeBSiC alloy ones (two-dimension connectivity) has near-ideal ME coupling. Very high ME effects of up to 22 V/cm Oe (4×10^{-7} s/m) at 1 Hz—an order of magnitude higher than the giant ones—have been found. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2420772]

Magnetoelectricity is the interaction between the polarization and magnetization subsystems of a solid. Early room temperature magnetoelectric (ME) measurements of Cr_2O_3 crystals[1,2] reported a value for the ME susceptibility of $\alpha_{ME}=2.67\times10^{-12}$ s/m, or equivalently a ME field coefficient of $\alpha_{ME}=\delta E/\delta H=0.01$ V/cm Oe, which remains to this day the highest value for a single phase material. Later, two-phase magnetostrictive/piezoelectric particulate composites in (0-3), (1-3), and (3-3) connectivity[3,4] and laminated composites in a (2-2) connectivity[5–14] were shown to have much higher ME effects. In particular, nonresonance giant ME field coefficients of up to $\alpha_{ME}=2$ V/cm Oe (or $\alpha_{me}\approx5\times10^{-8}$ s/m or C/m^2 Oe) have been reported for (2-2) (*L-T*) (longitudinally magnetized transversely poled) composites of piezoelectric Pb(Zr,Ti)O_3 (PZT) layers laminated with magnetostrictive $Tb_{1-x}Dy_xFe_{2-y}$ or Terfenol-D,[5] Permendur,[6] Fe–Ga,[7] or $NiFe_2O_4$ (Ref. 8) ones, which are orders of magnitude higher than those of single phase systems. Such giant values of α_{ME} occur when the magnetostrictive layer are in a dc biased piezomagnetic state near $H_{dc}\approx400-500$ Oe. In said laminates, the ME effect is a product tensor property—and not intrinsic to individual layers—combining the magnetoelastic and elastoelectric effects of individual layers, via an elastic coupling between layers: an applied H produces an elastic strain in the magnetostrictive phase that is stress coupled to that of the piezoelectric one, resulting in an induced voltage.

Theoretical investigations[9,10] have shown that two-phase laminates in the (*L-L*) configuration should have yet significantly higher values of α_{ME}—one to two orders of magnitude—than that which has been experimentally reported.[10] In fact, theoretically, the (L-L) mode should have by far the highest values of α_{ME} than any other. The theoretical limit of $\alpha_{ME}^{(L\text{-}L)}$ (in V/cm Oe) in the case of perfect or ideal ME coupling is given as

$$\alpha_{ME}^{(L\text{-}L)}=\frac{nd_{33,m}g_{33,p}}{ns_{33}^{E}(1-k_{33}^{2})+(1-n)s_{33}^{H}}\quad(0<n<1),\qquad(1)$$

where s_{33}^E and s_{33}^H are the elastic compliances for the piezoelectric and magnetostrictive layers, k_{33} the electromechanical coupling coefficient of the piezoelectric layer, $d_{33,m}$ and $g_{33,p}$ the longitudinal piezomagnetic- and piezoelectric-voltage coefficients, and n a thickness fraction of magnetostrictive layers. Equation (1) neglects (i) the demagnetization factor's effect due to our long-type construction and (ii) the direct effect of the magnetic permeability (μ) which is indirectly accounted for by the effective piezomagnetic coefficient: $d_{33,m}=\mu_{33}^{S}s_{33}^{H}\lambda_{33}$, where λ_{33} is the magnetostriction coefficient. Equation (1) predicts a large α_{ME} of ~10 V/cm Oe for a (*L-L*) Terfenol-D/PZT ME laminate (for $n=0.5$), which is in significant discrepancy with the experimentally observed value of $\alpha_{ME}=0.5$ V/cm Oe [in our previous reports,[10] a rectifying factor β (in N/V) was empirically introduced for Eq. (1) adjustment]. Clearly, in spite of the significant progress made in the discovery of giant ME effects,[5–12] only a fraction of the potentially available magnetoelectricity has been released for the (*L-L*) configuration.

However, in prior reports, several important things had not been thoroughly considered that have significant ramifications on the encroachment of $\alpha_{ME}^{(L\text{-}L)}$ upon this theoretical limit. First, the giant magnetostriction in ferromagnetic materials (Terfenol-D, Fe–Ga, etc.) used in prior studies had quite low relative magnetic permeabilities: $\mu_r<10$. This results in correspondingly low effective piezomagnetic coefficients and larger required magnetic biases: it is $d_{33,m}$ (i.e., $d\lambda/dH$) and not magnetostriction λ that matters. This fact,

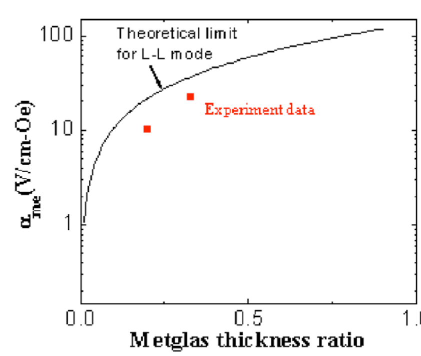

FIG. 1. (Color online) Graph of theoretical limitation of $\alpha_{ME}^{(L\text{-}L)}$ as a function of thickness fraction (n) of the magnetostrictive layers, calculated using Eq. (1) and shown as a solid line. Experimental data points are also shown in this investigation using piezofiber/Metglas (*L-L*) laminates. Note that unlike the *L-T* mode, when $n\to1$ for the *L-L* construction, $\alpha_{ME}^{(L\text{-}L)}$ does not tend to zero; rather, it tends to a maximum. This is because for the *L-L* mode, the polarization is along the length (or longitudinal axis) of the laminate, and thus with decreasing layer thickness the force imposed by a magnetostrictive layer of constant thickness increases.

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

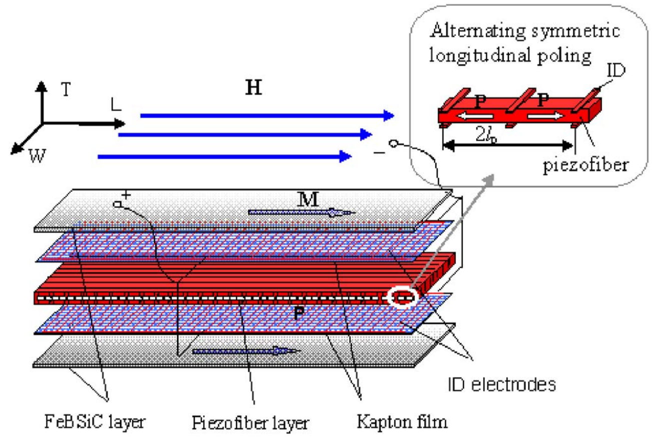

FIG. 2. (Color online) Illustration of our FeBSiC/piezofiber (L-L) laminate configuration. It consists of a 1D piezoelectric active fiber/epoxy composited (AFC) thin layer where the fibers are oriented along the longitudinal axis, which is laminated between two 2D FeBSiC layers. Insulating Kapton films with interdigitated (ID) electrodes were placed between layers. Each piezofiber had numerous alternating symmetric longitudinally poled "push-pull" units) that were each $2l_p = 1$ mm in length, as shown in the inset.

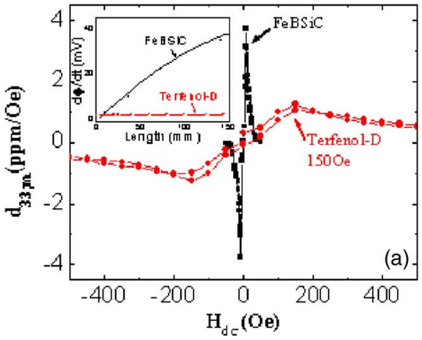

FIG. 3. (Color online) Effective piezomagnetic coefficients as a function of dc magnetic bias H_{dc} for ferromagnetic FeBSiC and Terfenol-D alloys. The inset shows the magnetic flux ($\delta\phi/\delta t$) as a function of the length of the ferromagnetic layer, for both single FeBSiC (cross-sectional area: $A = 7 \times 0.025$ mm^2) and Terfenol-D ($A = 6 \times 1.5$ mm^2) layers.

even though previously well known as given by Eq. (1), was not fully appreciated. Second, and more importantly, following Eq. (1), $\alpha_{ME}^{(L\text{-}L)}$ is proportional to the piezoelectric-voltage $g_{33,p}$ coefficient, rather than piezoelectric constant: $d_{33,p} = g_{33,p}\varepsilon_{33}$, where ε_{33} is the permittivity. This fact favors lamination of piezofibers [one-dimension (1D) phase connectivity] in a composite layer, rather than a monolithic piezoelectric one, to magnetostrictive layers. Early investigations of (1-3) piezocomposites were performed by Newnham *et al.*[13] Recently, investigations of (1-3) ME composites consisting of PZT rods in a Terfenol-D matrix were reported by Shi *et al.*[14] but unfortunately α_{ME} was <0.5 V/cm Oe under a required H_{dc} of 2000 Oe.

Recently,[13] we reported that high-permeability FeBSiC alloy exhibits large piezomagnetic coefficients $d_{33,m}$ at quite low dc magnetic biases and a large ME coefficient when bonded to a polyvinylidene fluoride piezopolymer layer. Here, we report a (L-L) ME composite consisting of a 1D phase connected piezoelectric PZT-fiber layer laminated between two two-dimension (2D) phase connected high-permeability magnetostrictive FeBSiC alloy (Metglas) foils, forming a magnetoelectric laminate with a (2-1) phase connectivity. This laminate has experimental values of $\alpha_{ME}^{(L\text{-}L)}$ approaching that of the theoretical limit calculated using Eq. (1), as shown in Fig. 1 by solid red points. Our findings demonstrate the unleashing of near-ideal magnetoelectricity latent in the (L-L) configuration.

Figure 2 illustrates our new (L-L) configuration, which consists of a piezofiber layer laminated between two high-permeability FeBSiC alloy ones. The FeBSiC layers were obtained from Metglas Inc. (Conway, SC), were foils of 25 μm thickness, had a relative permeability of $\mu_r > 40\,000$ due to a low magnetocrystalline anisotropy, and had a low saturation magnetostriction of $\lambda_s \approx 40$ ppm at $H_{dc} < 10$ Oe. The use of a thin-foil form offers the additional advantage of reducing eddy current losses at high frequencies. The piezo-fibers were PZT-5A ceramic ones that were 100 μm in thickness, 350 μm in width, and 30 mm in length. A (1-3) piezoelectric active fiber/epoxy composite (AFC) thin layer was then made, following a construction used in actuators.[16] The piezofibers were oriented along the longitudinal axis of the laminate. Thin polymer (Kapton) insulating films with interdigititated (ID) electrodes were placed between PZT-fiber and FeBSiC layers, which were then assembled into a laminated composite using an epoxy resin. Each piezofiber had numerous alternating symmetric longitudinally poled "push-pull" units with a length of $2l_p = 1$ mm, as shown in the inset of Fig. 2. This multiple push-pull (L-L) configuration not only optimizes stress transfer,[17,18] but also enhances the dielectric capacitance of the laminate. A second longer laminate was also constructed by laminating a (1-3) piezofiber layer (30 mm in length) between two long FeBSiC layers (100 mm in length). Following the composite nomenclature of Newnham *et al.*,[13] these ME laminates have a (2-1) connectivity of magnetic-piezoelectric phases.

In Fig. 3, we show the effective $d_{33,m}$ as a function of H_{dc} for ferromagnetic FeBSiC and Terfenol-D alloy layers. These values were calculated from the slope of striction-magnetic field (ε-H) curves measured using a strain-gauge method. It can be seen that the maximum value of $d_{33,m}$ is ~ 4 ppm/Oe under $H_{dc} \approx 10$ Oe for Metglas; whereas that of Terfenol-D was ~ 1 ppm/Oe under $H_{dc} \approx 200$ Oe. Although Terfenol-D has much higher magnetostrictions λ, its effective $d_{33,m}$ is notably lower than that of a FeBSiC alloy. This difference reflects the much higher magnetic permeability of FeBSiC: a high μ_r results in a low saturation field, and thus a large value of $d\lambda/dH$ at low H_{dc}. In addition, high-μ FeBSiC layers also concentrate magnetic flux ($d\phi/dt$). The inset of Fig. 3 shows $d\phi/dt$ as a function of the length (l) of FeBSiC and Terfenol-D layers, measured using a search coil. We found that the flux in FeBSiC layer (cross-section area is $A = 7 \times 0.025$ mm^2) is much higher (~ 20 times) than that of Terfenol-D ($A = 6 \times 1.5$ mm^2), and also that $d\phi/dt \sim l$ for the FeBSiC layer. These results demonstrate the potential of FeBSiC as a superior dc-biased piezomagnetic layer for ME laminate composites, one that can also operate under much reduced H_{dc}.

We then measured the voltage induced across the ID electrodes of the piezofiber layer as a function of (i) H_{dc} in response to a constant ac ($f = 1$ kHz) magnetic drive of $H_{ac} = 1$ Oe, both applied along the length of the laminate, and (ii) f under constant $H_{ac} = 1$ Oe and $H_{dc} = 4$ Oe. An electromagnet was used to apply H_{dc}, a pair of Helmholtz coils was used to generate a small H_{ac}, and the induced voltage was measured by a lock-in amplifier. First, in Fig. 4(a), we show $\alpha_{ME}^{(L\text{-}L)}$ as a function of H_{dc}. Data are given for both short ($l = 30$ mm)

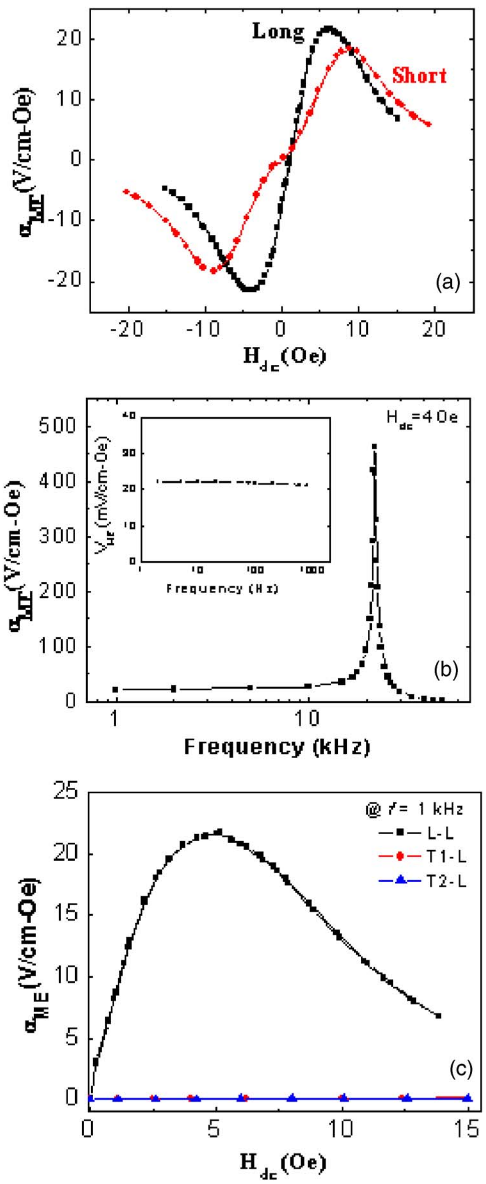

FIG. 4. (Color online) Magnetoelectric characterizations of FeBSiC/piezofiber laminates: (a) the (*L-L*) ME voltage coefficient $\alpha_{\rm ME}^{(L\text{-}L)}$ as a function of $H_{\rm dc}$ for both short (30 mm) and long (100 mm) laminates; (b) $\alpha_{\rm ME}^{(L\text{-}L)}$ as a function of frequency illustrating a strong enhancement at the electromechanical resonance frequency, where the inset shows a flat response over the quasistatic frequency range; and (c) anisotropy of $\alpha_{\rm ME}$ for $H_{\rm ac}$ applied along the length ($\alpha_{\rm ME}^{(L\text{-}L)}$), width ($\alpha_{\rm ME}^{(T1\text{-}L)}$), and thickness ($\alpha_{\rm ME}^{(T2\text{-}L)}$) of the laminate.

and long (l=100 mm) laminates, demonstrating that (i) ME coupling in FeBSiC/piezofiber laminates is much higher than that in Terfenol-D/monolithic-piezoelectric laminates; and (ii) $\alpha_{\rm ME}^{(L\text{-}L)} \sim l$ due to magnetic flux concentration effect, as shown in Fig. 3, which results in stronger magnetic induction in ME laminates. These data reveal maximum $\alpha_{\rm ME}^{(L\text{-}L)}$ values of 18 (short) and 22 V/cm Oe (long) under $H_{\rm dc}$ of 8 and 4 Oe, respectively: correspondingly $\alpha_{\rm ME}^{(L\text{-}L)} = 3.2\times10^{-7}$ and 4×10^{-7} s/m or C/m^2 Oe. These maximum values are much higher than recently reported *giant* ones ($\alpha_{\rm ME}^{(L\text{-}T)} \sim 0.5-2.2$ V/cm Oe,[5–8] $\alpha_{\rm ME}^{(L\text{-}L)} \sim 0.5$ V/cm Oe;[10,11] $H_{\rm dc}$ =300 Oe) and require much lower biases (<60 times). These values of $\alpha_{\rm ME}^{(L\text{-}L)}$ approach the theoretical limiting ones for the case of near-ideal ME coupling given by Eq. (1) (with n =0.33), as summarized in Fig. 1. These very high effects occur due to (i) optimum stress transfer in the (*L-L*) (2-1) configuration, (ii) the large $d_{33,m}$ and high permeability of the FeBSiC alloy layer, and (iii) the high $g_{33,p}$ of the 1D piezofiber layer. Theoretically, further enhancements in ME coupling are possible by increasing the magnetic phase ratio n (see Fig. 1).

Next, in Fig. 4(b), we show $\alpha_{ME}^{(L\text{-}L)}$ (l=100 mm) as a function of frequency under a constant bias of $H_{\rm dc}$=4 Oe. The inset demonstrates that very high values are maintained down to quasistatic frequencies. The figure itself reveals a strong resonance enhancement of $\alpha_{ME}^{(L\text{-}L)}$ to ~500 V/cm Oe at $f_r \approx 22$ kHz or, correspondingly, $\alpha_{\rm ME}^{(L\text{-}L)} = 10^{-5}$ s/m. Finally, in Fig. 4(c), we show $\alpha_{\rm ME}-H_{\rm dc}$ for $H_{\rm ac}$ applied along the length ($\alpha_{\rm ME}^{(L\text{-}L)}$), width ($\alpha_{\rm ME}^{(T1\text{-}L)}$), and thickness ($\alpha_{\rm ME}^{(T2\text{-}L)}$) of the laminate. The data show that $\alpha_{\rm ME}^{(L\text{-}L)}$ is dramatically larger than either $\alpha_{\rm ME}^{(T1\text{-}L)}$ or $\alpha_{\rm ME}^{(T2\text{-}L)}$, with a large anisotropy factor of $100 < K = \alpha_{\rm ME}^{(L\text{-}L)}/\alpha_{\rm ME}^{(T\text{-}L)} < 1000$, which can be attributed to the unidirectional natures of the (i) length-strain sensitivity of 1D piezofibers and (ii) demagnetization factor N of Metglas ribbon.

In summary, we have unleashed near-ideal magnetoelectricity latent in (2-1) laminates, consisting of a 1D connected piezoelectric PZT-fiber layer laminated between two 2D connected high-permeability magnetostrictive FeBSiC foils. Specifically, we find very high ME coefficients of $\alpha_{\rm ME}^{(L\text{-}L)} > 20$ V/cm Oe under small biases of 5 Oe, approaching predicted theoretical limits; (i) that are frequency independent in the quasistatic range; (ii) whose values are strongly enhanced, $\alpha_{\rm ME}^{(L\text{-}L)} \sim 500$ V/cm Oe, at the resonance frequency; and (iii) whose response is highly anisotropic, offering unidirectional sensitivity.

This work was supported by the Office of Naval Research and DARPA.

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