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Multi-electrode Pb(Zr,TiO)$_3$/Ni cylindrical layered magnetoelectric composite

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Multi-electrode Pb(Zr,TiO)$_3$/Ni cylindrical layered magnetoelectric (ME) composites were made by electroplating. The electroplated Ni layers were arrayed as four arcs on the inner PZT cylinder surface. The axial ME voltage coefficient of the composites was studied. Due to the cylinder symmetry, each of the four units of the PZT/Ni cylinder showed the same ME voltage response as the whole cylindrical ME composite, or when connected in parallel. When the four units were connected in series, the ME voltage was improved about three times than the single unit. This optimization is promising for the miniaturized ME devices design. © *2015 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4906614]

The magnetoelectric (ME) effect is the induction of a polarization by a magnetic field and/or a magnetization by an electric field in the ME materials, which has promising applications in novel multifunctional devices.[1–3] Recently, laminated ME composites have been formed by combining piezoelectric and magnetostrictive layers together, attracting great scientific interest due to their giant ME effect at room temperature. In such laminates, the ME effect arises from strain coupling across piezoelectric and magnetostrictive interfaces. To achieve larger ME effect, the interfacial bonding must be adequate to strengthen the coupling.[4,5] Pan *et al*.[6–8] demonstrated that Ni/PZT and Ni/PZT/Ni cylindrical ME laminates made by electroplating have giant ME coupling, owing to the rigid contact at the interfaces and the composite geometry.

Recently, several studies have concentrated on the ME effect in combined structures of electrically connected ME laminates/cantilever beams with different dimensions in series and/or in parallel. This way, both the ME voltage coefficient value and the resonance frequency can be modified, suitable for applications as magnetic field sensors and energy harvesters.[9–14] However, introducing more laminates is disadvantageous to the device miniaturization. Usually, the ME effect is characterized as the average response from the whole surfaces of the samples.[4–14] It has been theoretically and experimentally proven that different positions on laminated ME composites have different ME coupling, attributed to the different resonance modes and the corresponding strain distribution. All-surface electrodes on the piezoelectric layer produce an averaged ME effect and reduce the ME voltage, which is also true for the electrically conducting magnetostrictive layer serving as the electrode. This suggests that electrodes should only be deposited at the positions with higher ME response.[15,16] Nevertheless, few studies have reported circuit optimization of the electrodes on a single laminate. Based on these facts, PZT/Ni cylindrical layered ME composites with quartering electroplated Ni arc layers, which also serve as the positive electrodes, were designed. Considering the cylinder symmetry, the axial ME coupling of the four individual units was studied when connected in series or in parallel.

Two different structures of PZT/Ni cylinders were prepared by electroplating, as shown in Fig. 1. The two commercial PZT ceramic cylinders with the dimensions of $\Phi 25 \times \Phi 23 \times 10\,mm^3$ were polarized radially after electroless deposition of a thin Ni layer on its inside and outside surfaces as electrodes. Prior to electroplating Ni as the magnetostrictive layers, the two samples were pretreated with supersonic cleaning. The Ni electroplating bath composition and conditions are described elsewhere.[6] For the second sample, both positive and negative electrodes of the PZT cylinder were quartered by very narrow gaps along the cylinder axis to electrically isolate the electrodes, while the PZT cylinder remained intact. Then the four equally spaced positive electrodes were electroplated with Ni layers simultaneously. Since the PZT cylinder is an insulator, four ME measurement units (units 1, 2, 3, and 4) were classified by the negative and corresponding positive electrodes. Both samples have approximately the same magnetostrictive thickness of 340 μm.

The ME voltage coefficient measurements were performed in the ME measurement system, where both constant (H_{DC}) and alternating (δH) magnetic fields were applied parallel to the cylinder axis (axial mode). The ME voltage coefficient was calculated as $\alpha_{E,A} = \delta V/(t_{PZT} \cdot \delta H)$, where t_{PZT} is the PZT thickness and δH is the amplitude of the AC magnetic field generated by the Helmholtz coils.[6,7]

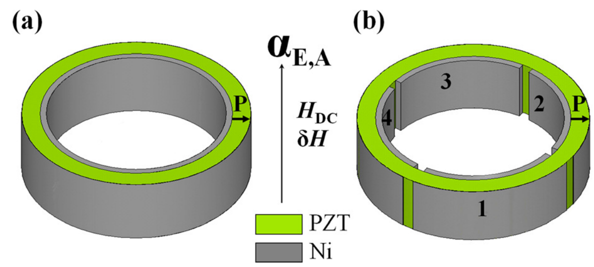

FIG. 1. Schematic illustration of (a) PZT/Ni cylinder; (b) PZT/Ni cylinder with arrayed Ni arc layers.

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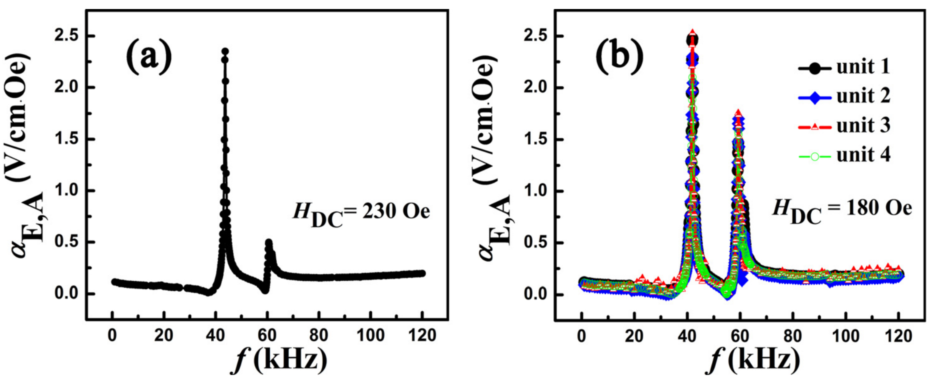

FIG. 2. The ME voltage coefficient, $\alpha_{E,A}$, dependence on the applied magnetic field frequency: (a) PZT/Ni cylindrical layered composite with H_{DC} = 230 Oe; (b) each of the four units of the PZT/Ni cylinder with quarter arrayed Ni arc layers with H_{DC} = 180 Oe.

The magnetic field frequency dependence of the ME voltage coefficient, $\alpha_{E,A}$, at the corresponding optimal DC magnetic field for the first sample and each of the four units of the second sample is presented in Fig. 2. Obviously, the first sample and the four units of the second sample have analogous ME voltage output under optimal H_{DC}. In all cases, there are two resonance peaks. The first and the second resonance peaks correspond to the axial and the radial electromechanical vibration resonance modes of the PZT cylinders, respectively.[8] The maximum $\alpha_{E,A}$ values are observed at the first peak, about 2.4 V/cm Oe. The results show that $\alpha_{E,A}$ will not be reduced, but rather almost unchanged when the Ni layer is divided into more sections. It is mainly because that the ME voltage coefficient is related to the properties of the piezoelectric and magnetostrictive layers, the interfacial coupling, and the thickness ratio between the two layers; however, the electrode area has slight effect on it.[17–19] Fig. 2(b) shows that the curves of the four units in the second sample overlap with each other and the two resonance $\alpha_{E,A}$ peaks of each unit appear at 42 kHz and 59.3 kHz. These facts demonstrate that the ME effect is almost equivalent for all the units. The negligible little differences in the ME effect of each unit may be attributed to the shape and the positioning errors during fabrication and measurements. As demonstrated by Pan *et al.*,[8] the cylindrical layered ME composite axial coupling can be simplified as a bilayer ME laminate longitudinal mode. The four units can be viewed as four quarters of the simplified laminate. In the axial mode, due to the cylinder symmetry, the size and the boundary conditions of the four units are identical. The effective magnetic field applied to the four Ni arc layers is along the cylinder's height and is uniform. Since the Ni layers and the PZT cylinder are rigidly bonded, they deform together. Thus, every unit has the same amount of stress-induced charge, denoted as Q_0. Zuo *et al.*[20] showed that each unit can be simply modeled as an LCR oscillator, thus only PZT sections are considered. Because of the same dimensions and applied magnetic field, each unit has the same capacitance, C_0. The ME voltage coefficient obtained from each unit is $\alpha^0_{E,A} = \delta V/(t_{PZT} \cdot \delta H) = \delta(Q_0/C_0)/(t_{PZT} \cdot \delta H)$. For the whole PZT/Ni cylinder, where both the stress-induced charge and the total capacitance are four times of the single unit, the ME voltage coefficient is $\alpha^W_{E,A} = \delta(4Q_0/4C_0)/(t_{PZT} \cdot \delta H) = \alpha^0_{E,A}$. The model predictions are consistent with the experimental observation that each unit has the same contribution to the ME voltage as the whole surface. It is notable that the ME charge coefficient ($\alpha_{Q,A}$ given as $\alpha_{Q,A} = Ct_{PZT} \cdot \alpha_{E,A}$)[21,22] is also the same for each unit due to the same capacitance, C_0, and PZT thickness, t_{PZT}. For the whole cylinder, $\alpha_{Q,A}$ is four times of the single unit as the capacitance is $4C_0$.

To obtain better ME performance from the second sample, the circuit was optimized by connecting the four units one by one in parallel and in series. Fig. 3(a) shows the maximum ME voltage coefficient $\alpha_{E,A}$ with the increasing number of the units connected either in series or in parallel. When the four units are connected in parallel, the electrodes on each PZT surface are shorted, just like the all-surface electrodes, so the maximum $\alpha_{E,A}$ remains at about 2.4 V/cm Oe, similar to the individual unit. However, the maximum $\alpha_{E,A}$ increases multiplicatively with the number of units connected in series. The maximum $\alpha_{E,A}$ is enhanced to about 7.2 V/cm Oe when all four units were connected in series, and the fitting line shows good linearity ($R^2 = 0.9910$). Compared with the whole PZT/Ni, or unit 1, the maximum $\alpha_{E,A}$ is improved by about three times. Meanwhile, the

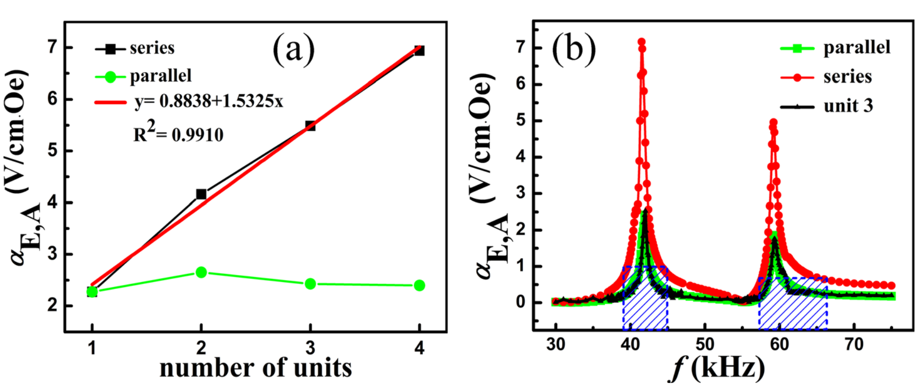

FIG. 3. (a) The maximum ME voltage coefficient, $\alpha_{E,A}$, as a function of the number of units connected either in series or in parallel; (b) the frequency dependence of $\alpha_{E,A}$ with H_{DC} = 180 Oe for the unit 3 alone and all the four units connected either in series or in parallel.

second peak $\alpha_{E,A}$ value exhibits a similar trend, not shown here. As for the mentioned model, when the four units are connected in series, the total capacitance of the circuit is reduced to $1/4C_0$, while the stress-induced charge remains unchanged at Q_0, so the total ME voltage is increased to $4\alpha^0_{E,A}$. With regard to the parallel connected mode, one can predict that the ME charge coefficient would increase linearly with the unit number m, since the capacitance is m times larger than the single unit, while the ME charge coefficient remains the same as for the single unit when the units are connected in series.[14] However, since in practice, the effect of mutual inductance among the four Ni layers and the non-harmonic ME response of the four units cannot be ignored,[23] the maximum $\alpha_{E,A}$ values are not merely added for the four units. Nevertheless, good linearity illustrates that the ME voltage output can be further improved when more electrodes (also the conducting magnetostrictive layer serves as an electrode) are divided and connected in series. Moreover, connecting ME array of m units in series not only increases the ME voltage output but also increases the signal-to-noise ratio by a factor of $\sqrt{m}$, which is helpful for the device design.[14,24]

Fig. 3(b) shows the comparison of the ME voltage output of the individual unit 3 and the four units connected either in series or in parallel over the 30 kHz to 70 kHz frequency range. The ME voltage coefficient is almost unchanged for the parallel connected mode, which confirms that the four units have the same ME voltage response. In contrast, for the series connected mode, since all the units exhibit the same resonant modes at the same frequency at the same time, overlapping of each peak contributes to the ME response enhancement by multiple times and broadens the frequency band, compared with the separate individual units. The frequency band where $\alpha_{E,A}$ is higher than 1 V/cm Oe is over 4 times wider than for the unit 3 alone at the first peak. In addition, the ME response in the frequency ranges far from the resonant band increased, compared with the single unit. Accordingly, without increasing the overall size, both the ME voltage coefficient and the bandwidth are optimized for the PZT/Ni cylinder with multiple electrodes connected in series.

In conclusion, a PZT/Ni cylindrical layered ME composite with quarter arrayed magnetostrictive Ni layer was fabricated and studied. Due to the symmetry of the cylindrical structure, the four Ni arc layers are equivalent under the axial magnetic field and the corresponding four units have the same ME voltage coefficient as the whole cylindrical ME composite, or when connected in parallel. By connecting the four array units in series, the ME voltage coefficient is enhanced multiplicatively with wider resonant band when the overall size is unchanged. This circuit optimization is helpful for the design of miniaturized ME devices, such as magnetic field sensors and energy harvesters.

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