

Magnetolectric resonance-bandwidth broadening of Terfenol-D/epoxy-Pb(Zr,Ti)O₃ bilayers in parallel and series connections

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The bandwidth of magnetolectric (ME) resonance for a combined structure where several Terfenol-D/epoxy-Pb(Zr,Ti)O₃ bilayers are connected in parallel and series was measured. A relationship between the bilayer length and resonance frequency was used to design the combined structure, in which the bilayered components of different lengths were connected in the parallel and series forms, respectively. The measured giant ME effect of the combined structure showed a much wider frequency response to external field than a single bilayered structure, and the ME effect far from the resonance ranges was enhanced significantly too. A qualitative analysis based on the equivalent circuit concept was presented to explain these effects. © 2005 American Institute of Physics. [DOI: 10.1063/1.1854736]

The magnetolectric (ME) effect is defined as a polarization P response to an external magnetic field H and vice versa.¹ This phenomenon is studied in both single-phase materials² and bulk piezoelectric/magnetostrictive composites.^{3,4} In the past few years, laminate or layered composites were prepared in order to achieve giant ME effect and ultrahigh sensitivity.⁵⁻¹⁰ For rectangular layered composites, such as Terfenol-D/Pb(Zr,Ti)O₃, several experiments reported an enhanced ME voltage coefficient (~ 10 Vp/Oe) at the resonance frequency.^{9,10} In the meantime, the calculation and modeling of the ME coupling in laminate composites were performed.^{8,11-13} Both experiments and theories predicted a sharp peak at resonant frequency and consequently an ultrahigh ME voltage coefficient.

The giant ME effect in layered composites lies in a mechanical stress-mediated electromagnetic coupling. Under an ac-electric field at resonance frequency, this coupling is most significant. The prominent ME transduce has to take place only in a very limited range near the resonant frequency. Moreover, the resonant point strongly depends on the dimension and shape of the resonator. Upon fluctuations of the operation conditions, resonance frequency of the device may change significantly, resulting in a decreasing of the device sensitivity, and thus the qualification of the devices. Our motivation is to develop a combined ME resonator which shows a much wider range of response frequency, i.e., a wide bandwidth.

An ideal design is to develop a combined structure of ME composites in which each component has slightly different resonance frequency, so that the effective resonance bandwidth can be widened. As the first step, the resonance frequency of each component must be accurately controlled. It is well known and also established in our experiments that for rectangular piezoelectric/magnetostrictive bilayered composite component, the characteristic resonance frequency is determined by its length if other parameters remain fixed. Empirically, we say, the one-dimensional approximation ap-

plies when the component length is four times its width. By this approximation, the resonator (component) has a "half-wavelength resonance," which can be strictly resolved from its motion equation. This relationship between the length and resonance frequency is well known in piezoelectricity and can be simply written as $f_r \times L = V/2 \equiv \text{constant}$, where f_r is the resonance frequency and L is the length of component, V is an average acoustic velocity of the laminate and is related to the thickness ratio of magnetostrictive and piezoelectric layers.¹¹ This inversely proportional relation allows us to argue that a combined structure consisting of several bilayered components of slightly different lengths may exhibit several resonance frequencies at the same time which are very close to each other. An overlap of these resonance states constitutes an output spectrum of a bandwidth wider than each of these components. Our experiments reported below follow this technical roadmap.

Our ME combined structure consists of a series of rectangular bilayered components connected in parallel and series forms, respectively. Each component is fabricated by one Pb(Zr,Ti)O₃ (PZT) piezoelectric ceramic layer polarized in thickness direction bonded with one Terfenol-D(Tb_{1-x}Dy_xFe_{1-y})/epoxy layer magnetized along its length direction to form a bilayered structure. PZT-502 (from PKI, USA) used for the piezoelectric layer was coated with silver electrodes and poled with 40 kV/cm electric field in 395 K silicone oil for 15 min after being sliced. The slices of Terfenol-D and PZT were 0.7 and 0.5 mm thick, respectively, and bonded with glue. We prepared four bilayered components with different lengths of $L=12.5, 13.5, 14.5, 15.5$ mm, all 3 mm wide and 1.2 mm thick. The sketch map of a ME rectangular bilayer is shown in Fig. 1. An ac sweep-frequency magnetic field (peak value 5 Oe) was applied along the length direction with a fixed and optimized bias magnetic field 700 Oe (the measurements were performed under different-frequency 0–3 kOe dc magnetic field and at 700 Oe the response is maximum). The induced ME voltage was measured between the top and bottom surface electrodes of PZT plate across the thickness direction.

First, we measured the ME coupling of the four bilayered components separately. A very sharp peak was obtained

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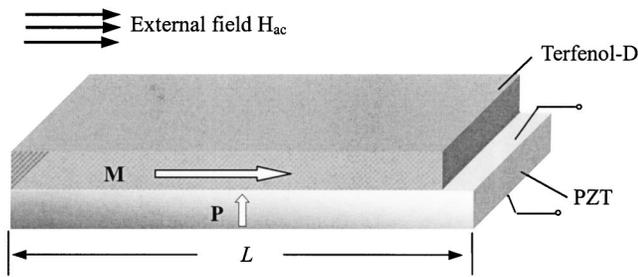


FIG. 1. Sketch diagram of a bilayer structure of Terfenol-D/PZT. The arrows P and M represent the polarization and magnetization direction, respectively. PZT is poled in the thickness direction and magnetic field is along length direction.

for each component at its resonance frequency, as shown in Fig. 2(a). The induced maximum ME voltages are 4.17–5.28 V responding to the applied magnetic peak value 5 Oe. For each of the four peaks, the 3 dB frequency bandwidth about resonant frequency f_r is determined as $\Delta f = f_2 - f_1$. Thus, although the resonant ME voltage is extremely high, the resonant bandwidth is very narrow. Consequently,

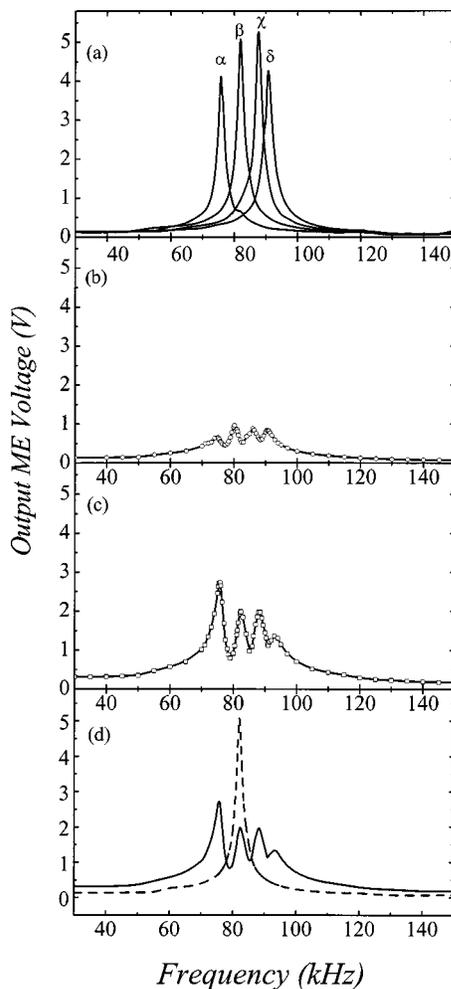


FIG. 2. Measured ME induced voltage vs frequency under an ac applied magnetic field with peak value 5 Oe, and a fixed bias magnetic field 700 Oe, both along length direction. (a) Resonance peaks of four bilayers, from left to right with different length: $\alpha \sim 15.5$ mm, $\beta \sim 14.5$ mm, $\chi \sim 13.5$ mm, $\delta \sim 12.5$ mm. (b) The case of P-connected structure. (c) The case of S-connected structure. (d) The band effect of the S-connected structure (solid line) in contrast with the resonance peak of the single-component sample with 14.5 mm length (dash line).

TABLE I. Some measuring and calculating results from Fig. 2(a).

Length of sample L (mm)	12.5	13.5	14.5	15.5
Resonance frequency f_r (kHz)	90.7	87.6	81.8	75.8
3 dB bandwidth Δf (kHz)	1.98	1.9	1.8	1.5
$L \times f_r$ (m/s)	1133.7	1182.6	1186.0	1174.9
Quality factor $Q_m(f_r/\Delta f)$	46	46	45	50

the effective mechanical quality factor of the laminate is $Q_m = f_r/\Delta f$. We also noticed that the resonant frequency has a blueshift as the length of the components decreases (from α to δ in the plots). This property can be well described by $f_r \times L \equiv \text{constant}$. The results are listed in Table I.

We developed two types of combined structures by connecting the four components one-by-one in the parallel (P-connected) and series (S-connected) forms, respectively. The measured output is the shunt voltage for the P-connected structure and series voltage for the S-connected one. Figure 2(b) shows the measured ME-induced voltage as a function of frequency for the P-connected structure. Indeed one observes four peaks which correspond, respectively, to the resonance of the four components. However, the pattern of the four peaks is obscure and an overlapping of the four peaks produces a broad band within which the magneto-electric coupling maintains a relatively high level. This broad band with high ME output is really what we prefer in this study. However, it is also evident that the maximum voltage of this broadened band is small compared to the effect recorded for the separate individual component, even smaller than 1 V.

Alternatively, the S-connected combined structure was attempted. The measured spectrum is shown in Fig. 2(c). Though the induced output is also smaller than the peak value for the separate individual component, the effective band is widened significantly and the data in the broadened resonance band are all above 1 V and the maximum even reaches 2.5 V. We again compare the output of the individual component and the S-connected structure in Fig. 2(d) where the dashed line is for the former and the solid line is for the latter. For the solid line, the regions of resonance are overlapped and the bandwidth for which the output voltage > 1 V is about 30 kHz. In addition, the ME effect recorded in those frequency ranges far from the resonant band becomes ~ 2 times of that for the single component. Therefore, the enhanced ME output over the whole frequency range is due to the accumulation effect of the S-connected combined structure. Obviously, the P-connected and S-connected combined structures both bring a broadening band but the latter structure shows more significant effect. Furthermore, the S-connected combined structure has enhanced ME output in the nonresonance ranges, in comparison to the single component.

Now we discuss the possible origin of the broadening effect of the resonance bandwidth for the combined structure. We present a simple equivalent-circuit modeling and a qualitative analysis on this effect. A magneto-elasto-electric bi-effect equivalent circuit of longitudinal and transverse ME laminate was developed in Ref. 11. However, due to the complexity of the problem for the two types of four component-connected structures, we only took the PZT piezoelectric layer into account when we deal with the output voltage. Simply assuming each piezoelectric resonator to be a LCR oscillator shown in Fig. 3(a) containing one inductor,

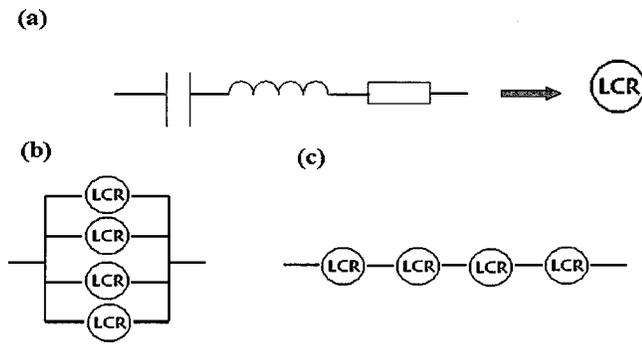


FIG. 3. A sketch map of simple LCR equivalent circuit. (a) An LCR oscillator circuit containing one capacitor, one inductor, and one resistance. (b) Parallel and (c) series connection of LCR to illuminate the voltage descent of the two combined structures.

one capacitor, and one resistance, we obtained the simple equivalent circuit of P- and S-connected of LCR in Figs. 3(b) and 3(c), respectively. Each LCR has the resonance frequency $\omega_p = 1/\sqrt{LC}$. In Fig. 3(a) and in the case of P-connected structure, the capacitors are parallel and the shunt capacitance decreases the voltage. If the four capacitors have equal capacitance, the output will drop to 1/4 of that for one capacitor. Consequently, the P-connected structure results in an obvious descent of the resonance peak. In the case of S-connected structure, as shown in Fig. 3(b), a series combination of capacitance does not bring a decrease of the voltage. However, in an LCR circuit, the impedance cannot be neglected. In the series circuit, the total impedance is $Z_s = \sum Z_i \approx 4Z_1$, where Z_i is the impedance of the i th ($i = 1-4$) LCR oscillator and they are approximately equal as the four laminates have almost the same length. The impedance induces the current and energy loss of the circuit and then results in reduction of output voltage. For the P-connected structure, the total impedance is $Z_p \approx Z_1/4 = Z_s/16$, so the parallel impedance effect is not as significant as the series one. In a word, the main cause of the reduced voltage for the P-connected structure is the shunt capacitance and for the S-connected one the main reason is from current loss due to the series impedance.

In conclusion, we have fabricated four Terfenol-D/epoxy-Pb(Zr,Ti)O₃ bilayered components with slightly dif-

ferent lengths, and designed the P-connected and S-connected combined structures, respectively, for obtaining enhanced ME performance. The measured results indicated a notable broadening of the resonance bandwidth in both combined structures, although the resonance peaked value is slightly lower than that for the individual component. It has been demonstrated that the S-connected structure shows a much higher resonance ME output and wider bandwidth than the P-connected one. In particular, a significantly enhanced ME output over the nonresonant frequency ranges for the S-connected structure has been demonstrated. An origin for this effect based on the simple LCR equivalent circuit approach has been presented.

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¹L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Gostekhizdat, Moscow, 1957).

²J. -P. Rivera and H. Schmid, *Ferroelectrics* **161**, 91 (1994).

³J. van Suchtelen, *Philips Res. Rep.* **27**, 28 (1972).

⁴J. Van den Boomgaard, A. M. J. G. van Run, and J. van Suchtelen, *Ferroelectrics* **14**, 727 (1976).

⁵J. Ryu, S. Priya, A. V. Carazo, and K. Uchino, *J. Am. Ceram. Soc.* **84**, 2905 (2001).

⁶J. Ryu, A. V. Carazo, K. Uchino, and H. E. Kim, *Jpn. J. Appl. Phys., Part 1* **40**, 4948 (2001).

⁷G. Srinivasan, E. T. Rasmussen, B. J. Levin, and R. Hayes, *Phys. Rev. B* **65**, 134402 (2002).

⁸M. I. Bichurin, D. A. Filippov, V. M. Petrov, V. M. Laletsin, N. Paddubnaya, and G. Srinivasan, *Phys. Rev. B* **68**, 132408 (2003).

⁹S. Dong, F. Bai, J. F. Li, and D. Viehland, *Appl. Phys. Lett.* **83**, 2263 (2003).

¹⁰S. Dong, J. Cheng, J. F. Li, and D. Viehland, *Appl. Phys. Lett.* **83**, 4812 (2003).

¹¹S. Dong, J. F. Li, D. Viehland, *IEEE Trans. Ultrason. Ferroelectr. Freq. Control* **50**, 1253 (2003).

¹²C.-W. Nan, *Phys. Rev. B* **50**, 6082 (1994).

¹³Y. X. Liu, J. G. Wan, J.-M. Liu, and C.-W. Nan, *J. Appl. Phys.* **94**, 5111 (2003).