Magnetoelectric Properties of a Heterostructure of Magnetostrictive and Piezoelectric Composites

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***Abstract*—This paper presents a magnetoelectric (ME) heterostructure of a magnetostrictive Terfenol-D/epoxy 0-3 composite (MSCP) plate combined with a piezoelectric PZT/epoxy 0-3 composite (PECP) plate along the longitudinal direction. By keeping the dimensions of the MSCP at 6.8 mm $\times$ 6.2 mm $\times$ 0.7 mm, heterostructures with various PECP lengths of 8–15 mm are prepared and characterized to reveal their frequency dependence of the ME effect. The results exhibit several resonance peaks in the heterostructures due to the magnetomechanical resonances (MMR) of the MSCP and the electromechanical resonances (EMR) of the PECP. The superposition of the MMR and EMR enhances significantly the ME output of the heterostructures. The highest ME voltage coefficient (α_E) of 13.1 V/cm · Oe is observed at the resonance frequency of 106.8 kHz when the length ratio of the MSCP and PECP is set to 6.8/10.2, together with an optimal magnetic bias field of 0.7 kOe.**

***Index Terms*—Heterostructures, magnetoelectric (ME) composites, magnetostrictive composites, piezoelectric composites.**

I. Introduction

THE magnetoelectric (ME) effect has received continuous attention due to its important application in transducers for conversion between magnetic and electric field signals. This effect is defined as a variation of dielectric polarization in a material when subjected to an applied magnetic field, or an induced magnetization in response to an external electric field [1]. Numerous ME single-phase materials have been reported since the discovery of the ME effect in antiferromagnetic Cr_2O_3 compound in 1961 [2]. However, none of them meet the requirement for practical applications owing to their low ME effect characterized by the ME voltage coefficient (i.e., $\alpha_E \sim$ 0.02 V/cm · Oe) [2]–[4]. By contrast, a much higher ME effect

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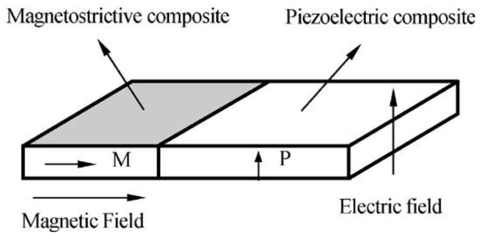

Fig. 1. Schematic diagram of the ME heterostructure. The arrows M and P show the magnetostrictive direction of the MSCP and the polarization direction of the PECP, respectively.

can be obtained in a two-phase composite structure by using the product-property of the magnetostrictive effect in a magnetostrictive material and a piezoelectric effect in a piezoelectric material. That is, a magnetic field applied to the two-phase structure results in a mechanical strain in the magnetostrictive phase that subsequently stresses the piezoelectric phase to generate an electric field proportional to the applied magnetic field. At present, several bulk and laminate ME composites have been developed. On the side of the bulk composites, this includes cobalt ferrite (CFO)/barium titanium ($BaTiO_3$), nickel ferrite (NFO)/$BaTiO_3$, etc., and on the side of the laminate composites, this comprises ferrite/lead zirconate titanate (PZT), Terfenol-D/PZT, etc. [5]–[9]. The reported α_E values are in the range of 0.06–4.68 V/cm · Oe.

We have recently developed an ME heterostructure by combining a magnetostrictive Terfenol-D/epoxy 0-3 composite (MSCP) plate with a piezoelectric PZT/epoxy 0-3 composite (PECP) plate along the longitudinal direction (Fig. 1) [10], [11]. The magnetostrictive direction of the MSCP is in its longitudinal direction, while the polarization direction of the PECP is along its thickness direction. The heterostructure operates in the longitudinal vibrational mode, producing a high α_E of 8.7 V/cm · Oe at the resonance frequency of 59.2 kHz through the transfer of acoustic energies between the MSCP and PECP.

In this paper, the frequency dependence of the ME effect in the heterostructure is investigated by varying the length ratio of the MSCP and PECP. Our goal is to obtain an improved ME effect in comparison with the previously reported heterostructure by optimizing the length ratio of the material phases.

II. Experiment

Terfenol-D/epoxy 0-3 composite and PZT-502/epoxy 0-3 composite were selected as the MSCP and PECP in the heterostructure, respectively, due to their tailorable properties. An additional benefit of using Terfenol-D/epoxy composite, as

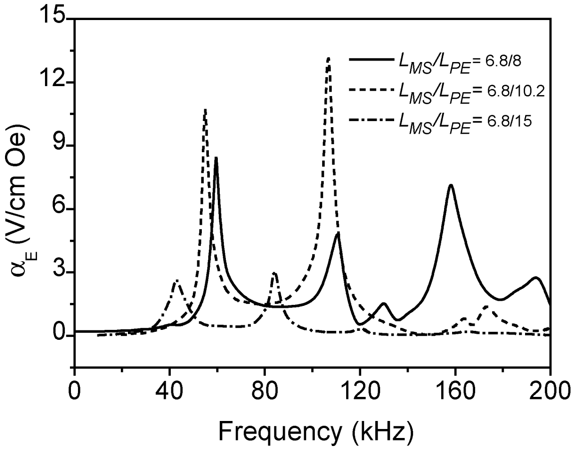

Fig. 2. Frequency dependence of α_E for heterostructures with various $L_{\rm MS}/L_{\rm PE}$ values and at an optimal magnetic bias of 0.7 kOe.

compared with bulk Terfenol-D, is its lower eddy-current losses at high frequencies [12]. The volume fractions of Terfenol-D (Baotou Rare Earth Research Institute, Inner Mongolia, China) and PZT-502 (Piezo Kinetics, Inc., Bellefonte, PA) in the MSCP and PECP were 0.7 and 0.6, respectively. The average sizes of Terfenol-D and PZT powders were 100 and 60 μm, respectively. The epoxy resin employed was Spurr low-viscosity embedding media (Polysciences, Inc., Warrington, PA). The heterostructure was prepared by using a cold compression technique. Detailed fabrication procedure can be found from a previous article [10]. In this study, the dimensions of the MSCP were fixed at 6.8 mm (length, $L_{\rm MS}$) $\times$ 6.2 mm $\times$ 0.7 mm, while those of the PECP were $L_{\rm PE}$ $\times$ 6.2 mm $\times$ 0.7 mm, where the length of the PECP ($L_{\rm PE}$) was set at 8, 10.2, and 15 mm.

The piezomagnetic coefficient of the MSCP ($d_{33,m}$) and the piezoelectric coefficient of the PECP ($d_{33,p}$) were measured to be 5.7 nm/A and 50 pm/V, respectively. The ME voltage coefficient (α_E) was determined by the electric field induced in the PECP under a small magnetic drive (or ac) field of 5 Oe (i.e., generated by a solenoid) superimposed onto a magnetic bias (or dc) field of up to 2.1 kOe (i.e., generated by a pair of permanent NdFeB magnets). Both the magnetic drive and bias fields were parallel to the longitudinal direction of the MSCP (or the heterostructure). The solenoid was driven by a signal generator (HP 33 120A) via a power amplifier (HSA 4014). The induced electric field was measured with a high input-impedance circuit connected to an oscilloscope (HP 45 845A). The magnetic drive field and magnetic flux density of the MSCP were, respectively, measured by a pick-up coil and a search coil connected to a fluxmeter (PF 900C). The electrical impedance of the PECP was measured by an impedance analyzer (Agilent 4294A).

III. Results and Discussion

Fig. 2 shows the frequency dependence of α_E for heterostructures with various $L_{\rm MS}/L_{\rm PE}$ values and at an optimal magnetic bias field of 0.7 kOe. The determination of this optimal magnetic bias field can be found elsewhere [10]. In all cases, no remarkable frequency dispersion is observed except for the resonance ranges, indicating an excellent frequency stability on the ME effect. For the heterostructure with $L_{\rm MS}/L_{\rm PE} = 6.8/8$, three significant resonances are observed at 59.2 kHz ($\alpha_E =$ 8.7 V/cm$\cdot$Oe), 110.3 kHz ($\alpha_E = 4.8$ V/cm$\cdot$Oe), and 157.7 kHz ($\alpha_E = 7.7$ V/cm$\cdot$Oe). In order to determine the physical origin of these resonances, frequency response measurements on both the magnetic flux density of the MSCP and electrical impedance of the PECP in the heterostructure were performed with the results being shown in Figs. 3 and 4, respectively. It can be seen from the heterostructure with $L_{\rm MS}/L_{\rm PE} = 6.8/8$ that the magnetomechanical resonances (MMR) of the MSCP appear at 59.1 and 109.8 kHz, while the electromechanical resonances (EMR) of the PECP occur at 60.8 and 156.2 kHz. Comparing these characteristic resonance frequencies with the α_E (or the ME) resonance frequencies obtained in Fig. 2, we find that the α_E resonance located at 110.3 kHz agrees with the second MMR of the MSCP at 109.8 kHz, whereas that situated at 157.7 kHz coincides with the second EMR of the PECP at 156.2 kHz. This indicates that the second and third α_E resonances are attributed to the second MMR of the MSCP and the second EMR of the PECP, respectively. On the other hand, the strongest α_E resonance detected at 59.2 kHz is in agreement with both the fundamental MMR at 59.1 kHz and EMR at 60.8 kHz, suggesting that the MMR superimposes partly the EMR. This resonance-superposition leads to a high α_E of 8.7 V/cm $\cdot$ Oe at 59.2 kHz.

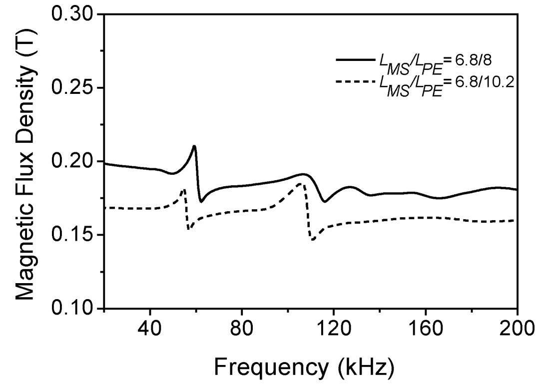

Fig. 3. Frequency dependence of the magnetic flux density for the MSCP in the heterostructures with various $L_{\rm MS}/L_{\rm PE}$ values and at 0.7-kOe bias.

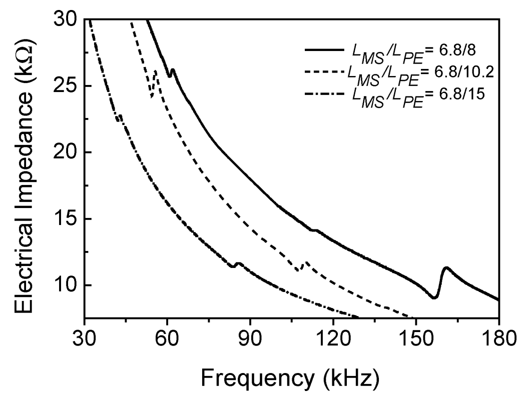

Fig. 4. Frequency dependence of the electrical impedance for the PECP in the heterostructures with various $L_{\rm MS}/L_{\rm PE}$ values and at 0.7-kOe bias.

It is interesting to note from the heterostructure with $L_{\rm MS}/L_{\rm PE} = 6.8/8$ that both the calculated magnetomechanical coupling coefficient of the second MMR (i.e., $k_{33,m} = 0.378$) and electromechanical coupling coefficient of the second EMR (i.e., $k_{33,p} = 0.245$) are larger than those of the fundamental resonances (i.e., $k_{33,m} = 0.311$ and $k_{33,p} = 0.188$) (see Figs. 3 and 4). Moreover, the α_E resonances at 110.3 and 157.7 kHz, caused by the second MMR and EMR, also have high values of 4.8 and 7.7 V/cm · Oe, respectively. Specifically, they are only slightly lower than the resonance-superposition-enhanced α_E of 8.7 V/cm · Oe at 59.2 kHz. Thus, it is possible to obtain a higher α_E by resonance-superposition of the second MMR and EMR instead of the fundamental MMR and EMR.

To obtain a deep insight into this resonance-superposition-enhanced effect, we have prepared an optimal heterostructure with $L_{\rm MS}/L_{\rm PE} = 6.8/10.2$. As shown in Figs. 3 and 4, the fundamental MMR and EMR occur at 54.5 kHz, while the second MMR and EMR appear at 106.8 kHz. They are in good agreement with the observed α_E resonances illustrated in Fig. 2, thus confirming that the MMR of the MSCP superimposes completely the EMR of the PECP. As shown in Fig. 2, the highest α_E of 13.1 V/cm · Oe is detected at 106.8 kHz. This α_E resonance, which corresponds to the second longitudinal vibrational mode of the heterostructure, is even higher than the fundamental mode located at 54.5 kHz (i.e., $\alpha_E = 11.3$ V/cm · Oe) and all the previously observed α_E resonances in the heterostructure with $L_{\rm MS}/L_{\rm PE} = 6.8/8$. It is likely that the whole heterostructure vibrates in the second longitudinal mode with its length $(L_{\rm MS} + L_{\rm PE})$ equal to one longitudinal wavelength. In more detail, $L_{\rm MS}$ and $L_{\rm PE}$ correspond to 0.5 longitudinal wavelength each.

Similar studies were also performed on a heterostructure with $L_{\rm MS}/L_{\rm PE} = 6.8/15$. The α_E resonances seen at 42.7 and 84.1 kHz (Fig. 2) match reasonably well the first and second EMR (Fig. 4). However, their amplitudes, 2.7 V/cm · Oe at 42.7 kHz and 3.1 V/cm · Oe at 84.1 kHz, are not as strong as the previous two heterostructures. We were unable to obtain the magnetic flux density response for the MSCP because of very low SNR. Nevertheless, the observation provides an indication that the use of a longer PECP reduces the actuation efficiency of the MSCP and thus leads to a decrease in the ME output of the heterostructure.

IV. Conclusion

Three ME heterostructures with different $L_{\rm MS}/L_{\rm PE}$ values have been fabricated by combining a MSCP plate with a PECP plate along the longitudinal direction. The ME effect of these heterostructures have been investigated as a function of frequency. By applying an optimal magnetic bias field of 0.7 kOe to the heterostructures, the highest α_E of 13.1 V/cm · Oe has been observed at the second resonance frequency of 106.8 kHz when $L_{\rm MS}/L_{\rm PE}$ is set to 6.8/10.2. This suggests that the second MMR of the MSCP superimposes completely the second EMR of the PECP, and the whole heterostructure vibrates in its second longitudinal mode. This resonance-superposition-enhanced α_E shows promising application of the proposed heterostructure in transducers for conversion between magnetic and electric field signals.

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