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Frequency response of magnetoelectric 1–3-type composites

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A three-phase magnetoelectric (ME) composite consisting of $Pb(Zr,Ti)O_3$ rods embedded in a matrix of Terfenol-D/epoxy (TDE) pseudo 1–3 composites has been fabricated. Besides the large ME effect, its frequency response under a magnetic bias field has been studied. It was found that the resonance shifts to lower frequency with increasing bias field. Due to the magnetomechanical characteristics of the TDE pseudo 1–3 medium, the composite shows a similar trend in ME performance. Magnetic-field dependence of the frequency shift provides a means to tune the performance of ME sensors based on the composite. © *2010 American Institute of Physics*. [doi:10.1063/1.3399703]

I. INTRODUCTION

Combining piezoelectric and magnetostrictive elements, magnetoelectric (ME) composites have attracted considerable interest recently. ME response is the phenomenon in which materials develop an electric field when subjected to an external magnetic field, or conversely, they exhibit magnetization change upon application of electric field. $Pb(Zr,Ti)O_3$ (PZT) is a widely used piezoelectric ceramics as it has superior piezoelectric properties.[1] Among rare-earth-iron alloys, Terfenol-D ($Tb_{0.3}Dy_{0.7}Fe_{1.92}$) is a well-known and popular magnetostrictive alloy which exhibits giant magnetostriction and low magnetic anisotropy at room temperature.[2] By combining the strong characteristics of the piezoelectric ceramics and magnetostrictive alloy, the composites have been found to exhibit large ME response.[3]

Similar to piezoelectric composites, the reported forms of the ME composites are classified according to the connectivity which is defined as the number of dimensions in which a phase is self-connected.[4] 0–3 composites consisting of piezoelectric and magnetic oxide particulates[5–8] and 2–2 laminated composites consisting of piezoelectric and magnetic oxide layers[9–11] are the common connectivity schemes reported previously. To overcome the mechanical and electrical weaknesses of the Terfenol-D material used in the diphasic ME composites, triphasic ME composites have been developed in recent years.[12–14]

Among various triphasic ME composites, pseudo-1–3-type ME composites have been found to give a large ME response which show a large potential for ME device applications.[15,16] The idea is similar to that of piezoelectric 1–3 composites which have been studied and developed for wide range of applications.[17–21] In this study, a triphasic ME composite consisting of PZT rods embedded in a pseudo 1–3 Terfenol-D/epoxy (TDE) matrix is reported. Frequency response of the ME composite under the magnetic bias field has been studied. The results give evidence to prove that coupling interaction exists between the PZT rods and TDE medium.

II. EXPERIMENT

PZT ceramic was used as the active piezoceramic phase due to its excellent piezoelectric properties. Terfenol-D particles (Gansu Tianxing Rare Earth Functional Materials Co., Ltd., China) were used as the magnetostrictive phase due to its giant magnetostrictive strain at both room temperature and low field. Araldite GY251/HY956 epoxy was chosen as the matrix of the pseudo 1–3 composite due to its low viscosity. The basic parameters of the materials used are listed in Table I.

PZT ceramic disks of 15 mm diameter and 1 mm thick were fabricated by dry pressing and sintering at 1305 °C for 2 h. The sample was poled in silicone oil along its thickness direction by applying a dc field of 4 kV/mm at 120 °C for 30 min. The electric field was maintained until the sample was cooled to 50 °C. After poling, the ceramic disks were short-circuited at 40 °C to remove the injected charges. For fabricating the mixture of Terfenol-D particles and epoxy, predetermined quantities of particles and epoxy were homogenously mixed and degassed under a vacuum to eliminate air bubbles.

The ME pseudo 1–3 composites were fabricated using a conventional dice-and-fill technique.[22] Poled ceramic disk was cut using a precision linear saw (Buehlet Isomet 4000) with a blade of 0.5 mm thickness. The ceramic width was around 0.4 mm so that the aspect ratio (thickness to width ratio) of the PZT rods inside the composites was higher than

TABLE I. Properties of PZT ceramic, Terfenol-D, and epoxy.

	PZT	Terfenol-D	Epoxy
Density (kg/m^3)	7700	9200	1100
Piezoelectric coefficient (pC/N)	400	⋯	⋯
Magnetostriction (ppm)	⋯	700	⋯
Young's modulus (GPa)	71	40	2.9

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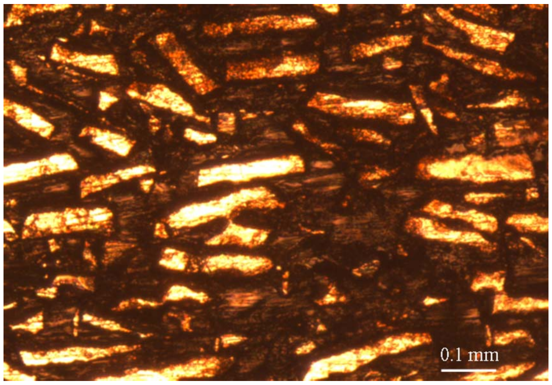

FIG. 1. (Color online) A microscopic picture of alignment of Terfenol-D particles embedded in the epoxy.

2 to avoid mode coupling. The well-mixed 32 vol % of Terfenol-D particles and epoxy slurry was filled into the grooves of the diced disk. The slurry-filled sample was further degassed in vacuum and subsequently placed between a pair of NdFeB permanent magnets which produces a uniform magnetic field of ~0.15 kOe along the thickness direction of the sample. The optical microscope was used to observe the alignment of the Terfenol-D particles embedded in the epoxy on the cross-section area of the composite. As shown in Fig. 1, the Terfenol-D particles were aligned inside the epoxy with the magnetic flux lines and formed in chains giving a kind of the pseudo 1–3 composite. After full curing of the epoxy, the composites were polished and then electroded with air dried silver paint before electrical characterization.

The impedance and phase of the samples were measured using an impedance/gain phase analyzer (Agilent 4294A). The dynamic magnetic field H_{ac} was provided by a Helmholtz coil driven by a dynamic signal analyzer (Ono Sokki CF5220) via a high speed power amplifier (NF Electronic Instruments 4025). The magnetic bias H_{Bias} was applied by a variable gap permanent magnet (Pasco EM-8641) (shown in Fig. 2). H_{ac} and H_{Bias} were measured by a pick-up coil and a Gaussmeter (F.W. Bell 7030), respectively. Sensitivity of the ME composite at different frequencies under different H_{Bias} was calculated using

$$\alpha_{33} = \frac{dE_3}{dH_3}, \tag{1}$$

where E_3 is the electric field generated in the thickness direction and $H_3 = H_{ac}$ is the applied ac magnetic field.

III. RESULTS AND DISCUSSIONS

A PZT ceramic/TDE pseudo 1–3 composite has been fabricated as shown in Fig. 3. The measured electrical impedance and phase versus frequency spectra of the bulk disk and the composite with electrode is shown in Figs. 4(a) and 4(b), respectively. Two main resonance peaks appear, which are associated with the planar and thickness mode resonances of the samples.[23] Compared to the bulk PZT ceramics, the harmonics of the planar mode resonance of the composite are very weak so that very pure planar and thickness resonances can be observed without a significant mode coupling.

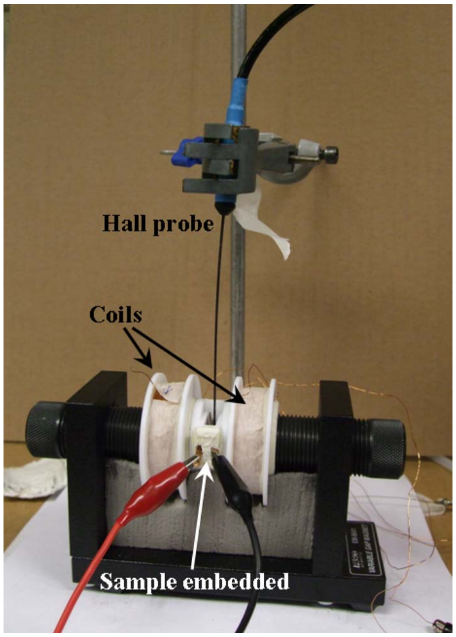

FIG. 2. (Color online) A photograph of the experimental setup for ME measurement.

Figure 5 shows a sample fixture for applying a magnetic bias in electrical impedance/phase measurements. A variable gap permanent magnet provides magnetic bias to the sample placed between them. A Hall probe (F.W. Bell STF71–0404–05), placed near the sample, was used to measure the magnetic bias generated by the magnets. Figures 6 and 7 show the variation in electrical impedance and phase of the planar mode resonance under different magnetic field H_{Bias}. Both the resonance peak and phase maximum shift to lower frequency with increasing magnetic bias. Figure 8 shows the dependence of the planar mode resonance frequency of the composite under the effect of different H_{Bias}. Initially, the resonance frequency decreases gently at low magnetic field. With increasing H_{Bias} into a high field region (~1.5 kOe), the resonance frequency drops significantly. This is due to the magnetomechanical and magnetostrictive characteristics of the TDE medium. The elastic modulus at constant magnetic field strength (Y_3) can be estimated using the resonance frequency by

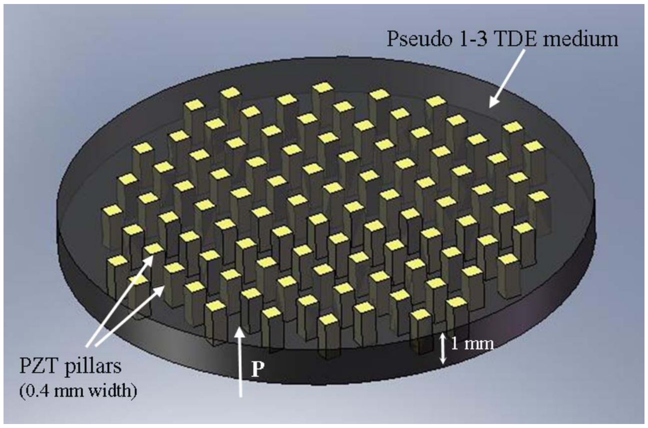

FIG. 3. (Color online) A schematic diagram of a ME pseudo 1–3 composite.

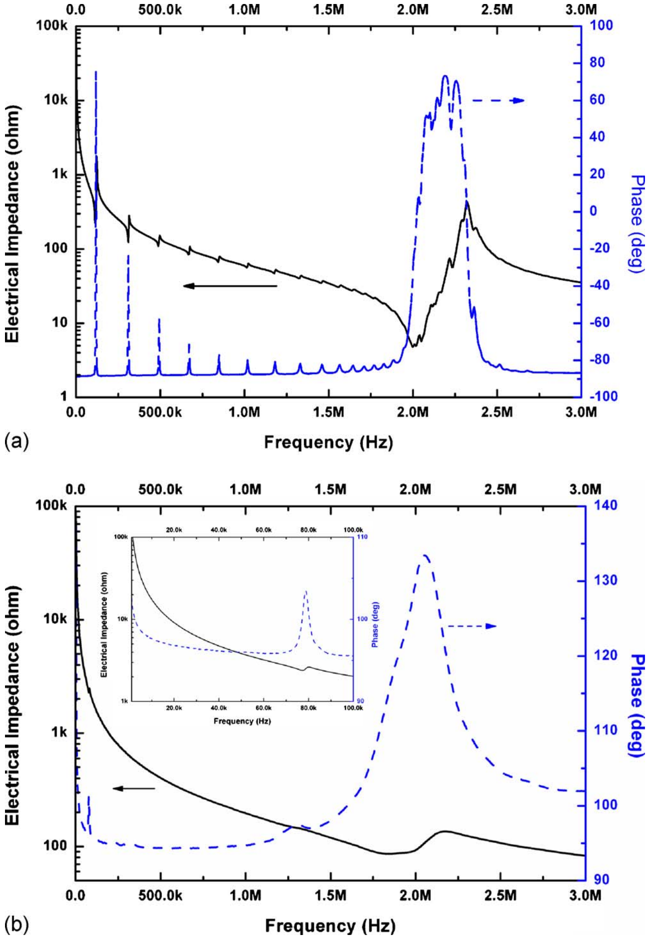

FIG. 4. (Color online) Electrical impedance and phase vs frequency spectra of (a) a PZT disk and (b) a ME pseudo 1–3 composite.

$$Y_3 = \rho(2tf_r)^2, \tag{2}$$

where t and ρ are the thickness and density of the composite, respectively. The TDE medium would show an initial drop in Y_3 with increasing H_{Bias}. As H_{Bias} is increased, Y_3 shows an increasing trend.[24] Since the volume fraction of Terfenol-D particles is low (ϕ=0.32), the increment of Y_3 would be still small even at a high H_{Bias} level. Meanwhile, the magnetostriction of the medium gets stronger with increasing H_{Bias} and then approaches to a peak at 2 kOe.[15] That is why the

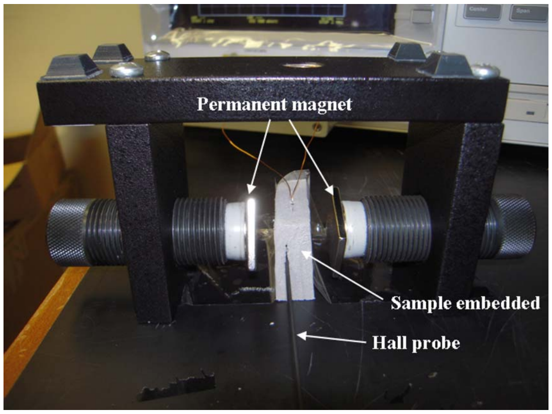

FIG. 5. (Color online) A sample fixture for applying a magnetic bias in electrical impedance/phase measurements.

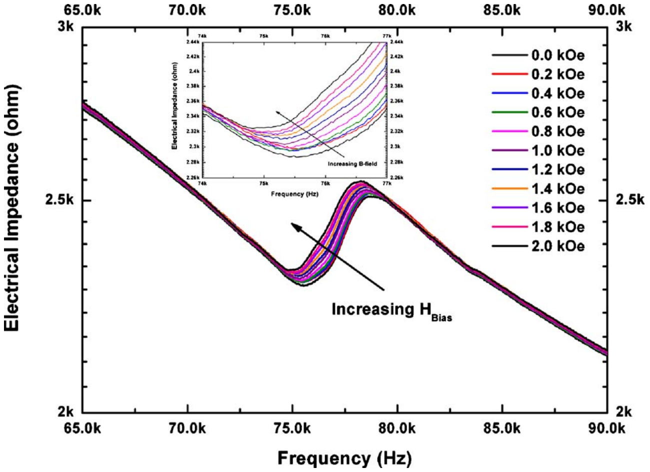

FIG. 6. (Color online) Electrical impedance vs frequency curves of a ME pseudo 1–3 composite as a function of H_{Bias} (inset: a magnified picture of resonance region).

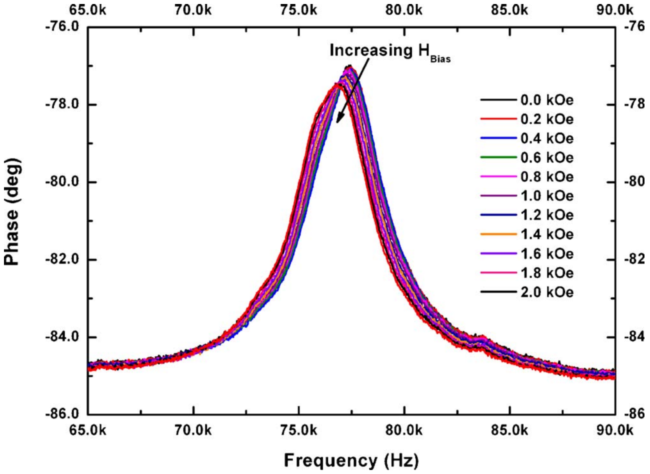

FIG. 7. (Color online) Phase vs frequency curves of a ME pseudo 1–3 composite as a function of H_{Bias}.

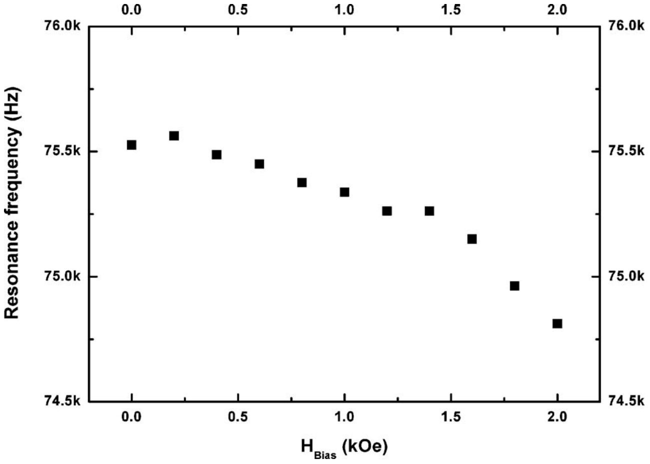

FIG. 8. Resonance frequency of a ME pseudo 1–3 composite obtained in electrical impedance measurement as a function of H_{Bias}.

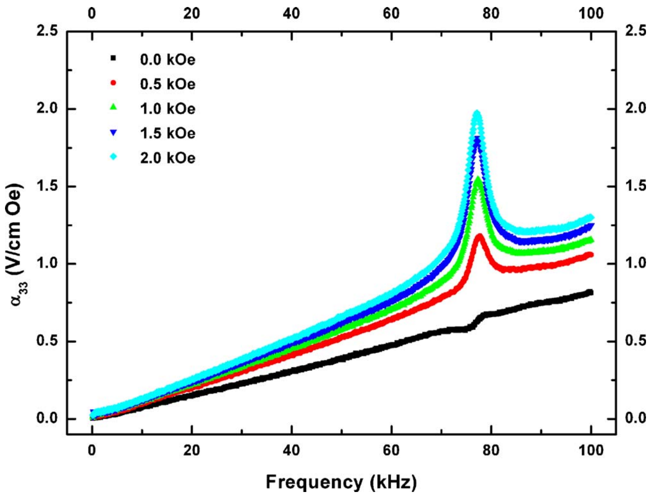

FIG. 9. (Color online) Frequency dependence of the ME coefficient as a function of H_{Bias} for the pseudo-1–3-type composite.

planar mode resonance frequency would decrease gently initially and then drop significantly at high magnetic field region.

Figure 9 shows the frequency dependence of the ME coefficient α_{33} of the composite under different H_{Bias}. As H_{Bias} increases, the ME resonance peak also shift to lower frequency (as shown in Fig. 10). The pattern is similar to that shown in Fig. 6 because the elastic properties of the TDE medium dominate the magnetic-field dependent ME performance of the composite.[25] Since the magnetic-mechanical-electric coupling between the TDE medium and PZT pillars is enhanced at resonance, a giant ME effect is found around the resonance region. The ME effect of ME 1–3-type composite at 1 kHz is shown in Fig. 11. It is seen that the ME coefficient α_{33} increases monotonically with increasing magnetic bias. At 2 kOe, the α_{33} value approaches 130 mV/cm Oe which is much higher than that reported for ME particulate composites ($\alpha_{33} < 50$ mV/cm Oe).[26–28]

IV. CONCLUSION

A ME PZT/TDE pseudo 1–3 composite has been fabricated using a conventional dice-and-fill method. The frequency response of the composite was studied using the impedance/phase measurement. It was found that the planar mode resonance frequency of the composite decreases gently initially under low magnetic field and then decreases significantly in the high field region. The trend is similar to that found in the ME measurement which presumably due to the magnetomechanical and ME characteristics of the TDE medium. Frequency shift in the ME composite provides a means to tune the sensitivity of the ME sensors in practical sensing applications.

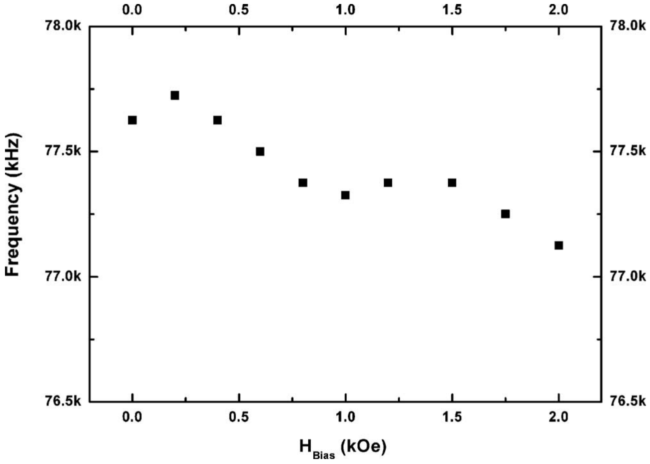

FIG. 10. Resonance frequency of a ME pseudo 1–3 composite obtained in ME measurement as a function of H_{Bias}.

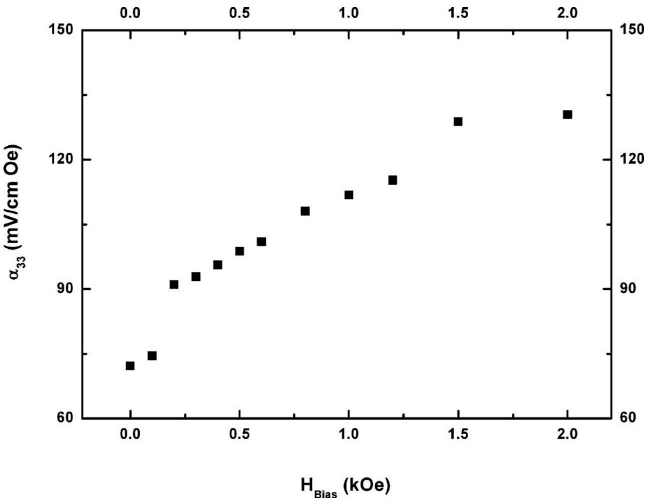

FIG. 11. ME coefficient α_{33} as a function of H_{Bias}, measured at 1 kHz for a ME pseudo 1–3 composite.

ACKNOWLEDGMENTS

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