Enhanced magnetoelectric effect in a stress-biased lead magnesium niobate-lead titanate single crystal/Terfenol-D alloy magnetoelectric sensor

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A stress-biased magnetoelectric sensor with a built-in permanent magnet has been fabricated using piezoelectric lead magnesium niobate-lead titanate single crystal and magnetostrictive Terfenol-D alloy. The resonance characteristics and magnetoelectric performance of the sensor have been evaluated under different stress-biased conditions. The resonance of the sensor shifts to higher frequency with increasing preloading stress. Due to the piezoelectric and magnetostrictive enhancements under preload, the device exhibits a giant magnetoelectric voltage coefficient of 0.22 V/Oe at a preloading stress of 2.5 MPa. This compact device has the potential to be used as a standalone sensor without requiring external power input. © *2011 American Institute of Physics*. [doi:10.1063/1.3536636]

I. INTRODUCTION

By combining the good performance of piezoelectric ceramics and magnetostrictive alloy, laminated magnetoelectric (ME) composites have been found to exhibit large ME responses.[1] ME performance of the composites depends significantly on the laminate configuration. Among various laminate configurations, the longitudinally poled longitudinally magnetized (LL) mode[2] and longitudinally magnetized transversely poled (LT) mode[3] have been used extensively to provide giant ME effects. Although it was reported that LT laminates show the highest ME effect experimentally, the LL laminates produce a strong stress coupling between layers.[4] However, in most of the published results, there are no mechanical stress bias applied on the ME composites or devices.

As known, domain walls move when subjected to an electric field or mechanical stress. Stress can induce non-180° switching which leads to a change in macroscopic strain.[5,6] Stress-induced non-180° domain switching has been reported in barium titanate and lead titanate single crystals. In barium titanate, 90° domains have been shown to nucleate under a specific level of compressive stress.[7] In Terfenol-D ($Tb_{0.3}Dy_{0.7}Fe_{1.92}$), a peak dynamic magnetomechanical strain coefficient can also be found at an optimum compressive stress.[8] It is because the application of an external compressive load can rotate domains to a direction perpendicular to the field axis (non-180° states), which is called magnetostrictive "jumping."[9,10] Since both magnetostrictive properties of the magnetostrictive material and piezoelectric performance of the piezoelectric material can be enhanced under certain optimal mechanical preload, the ME effect of the preloaded magnetostrictive/piezoelectric composite could also be enhanced. A recent study has reported that a stress-biased mechanism can enhance ME charge coupling in ME composites.[11] By combining a lead zirconate titanate

multilayer stack and a Terfenol-D rod, the ME charge coefficient of the composite under a specific preload is 100–1000 times higher than that previously reported for other ME laminates.

Besides piezoelectric ceramics, lead magnesium niobate-lead titanate (abbreviated as PMN-PT) single crystals with high piezoelectric coefficients and electromechanical coupling coefficients[12] have also been used as a piezoelectric phase in magnetoelectric composites. 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.[13] Giant ME effects in PMN-PT/Terfenol-D composites have been reported which show their potential in the applications of magnetic sensors and electromagnetic force transducers.[14,15]

In this paper, a ME sensor based on a stress-biased LL configuration of PMN-PT single crystal and Terfenol-D alloy has been studied. With a stable dc magnetic bias provided by a built-in NdFeB permanent magnet, the device can operate as a standalone sensor without requiring external power input. Through the piezoelectric and magnetostrictive enhancements under an optimum mechanical preload, a giant ME performance in the sensor is found.

II. EXPERIMENTS

PMN-PT single crystal with 28 mol % of PT content was grown in the Shanghai Institute of Ceramics by the modified Bridgman method.[16] The crystal was cut into a rectangular shape of 5×5 mm^2 area and 0.3 mm thick with the [001] orientation along the thickness direction. The sample was poled in silicone oil along its thickness direction by applying a dc field of 1 kV/mm at 130 °C for 10 min. The electric field was maintained until the sample was cooled to 50 °C. After poling, the single crystal plates were short-circuited at 40 °C to remove the injected charges. The [112]-oriented textured Terfenol-D alloy (Gansu Tianxing Rare

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TABLE I. Properties of PMN-PT single crystal and Terfenol-D.

	PMN-PT	Terfenol-D
Density (kg/m³)	7900	9200
Piezoelectric coefficient (pC/N)	1500	⋯
Magnetostriction (ppm)	⋯	700
Young's Modulus (GPa)	12	40

Earth Functional Materials Co., Ltd., China) rod has 4.5 mm diameter and is 12 mm long. The basic parameters of the materials used are listed in Table I.

Figure 1 shows a schematic diagram of the stress-biased ME sensor fabricated with the PMN-PT crystal and Terfenol-D alloy. The sensor consists of a Terfenol-D rod sandwiched between two thin PMN-PT single crystal plates, which are assembled into a stiff stainless steel frame. The Terfenol-D rod and PMN-PT single crystal plates are mechanically attached in series without using epoxy. A preloading stress is applied and can be adjusted using a bolt at the end of the frame. With this design, the Terfenol-D rod can be excited into a longitudinal motion under a magnetic field applied along the longitudinal-axis of the sensor. As a consequence, the single crystal plates, prestressed with the Terfenol-D rod, are forced to vibrate along the same direction. A small permanent magnet (~450 Oe) is installed at the end of the frame, which provides a dc magnetic bias to the PMN-PT single crystal/Terfenol-D sensor. With a built-in permanent magnet, the compact device can be used to detect magnetic field as a standalone sensor without requiring external power input.

The impedance and phase of the ME sensor were measured using a precision impedance analyzer (Agilent 4294A). The dynamic magnetic field H_{ac} was provided by a pair of Helmholtz coils driven by a dynamic signal analyzer (Ono Sokki CF5220) via a high speed power amplifier (NF Electronic Instruments 4025). H_{ac} was measured by a pick-up coil and a Gaussmeter (F.W. Bell 7030), respectively. The

FIG. 2. (Color online) Electrical impedance vs frequency spectrum of a PMN-PT single crystal/Terfenol-D ME sensor under different stress-biased conditions. [Inset: a magnified picture of resonance region]

magnetoelectric voltage coefficients at different frequencies under different H_{ac} are calculated using the following:

$$\alpha_{33} = \frac{dV_3}{dH_3}, \tag{1}$$

where V_3 is the voltage generated on the PMN-PT surface by the vibration of the Terfenol-D rod, and $H_3 = H_{ac}$ is the applied ac magnetic field. The ME measurement of the sensor was performed under free and different stress-bias conditions.

III. RESULTS AND DISCUSSIONS

The frequency-dependent electrical impedance and phase of the ME sensor under different stress-biased conditions are shown in Figs. 2 and 3, respectively. It can be seen that both the resonance peak and phase maximum shift to higher frequency with increasing preloading stress. This can be explained by the following equation:[17]
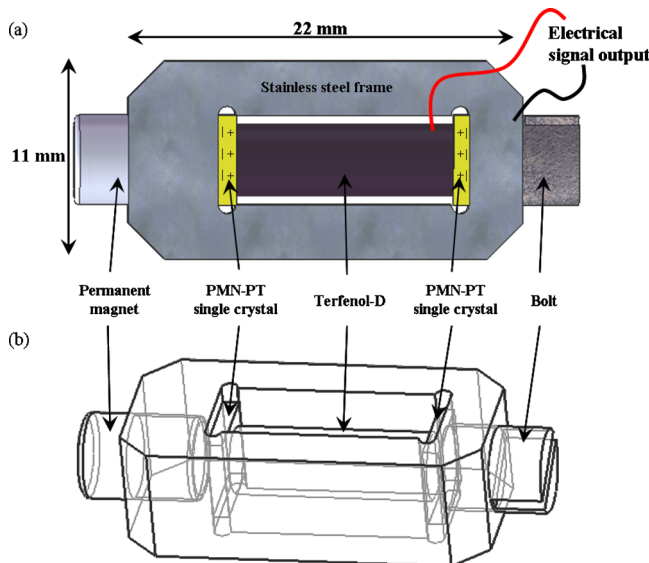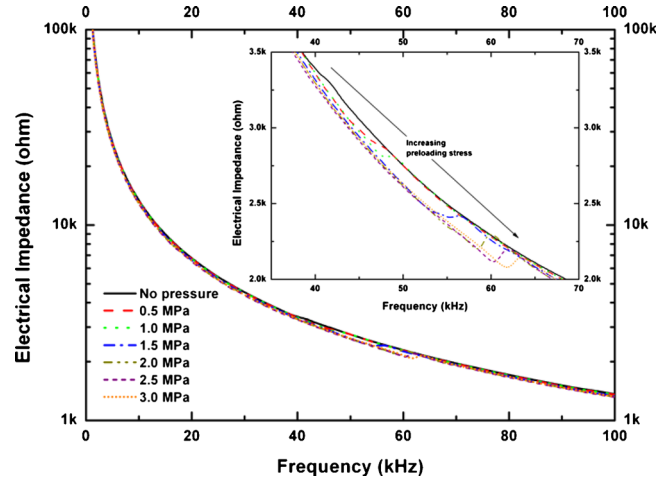

FIG. 1. (Color online) A schematic diagram of (a) top view and (b) isometric view of a stress-biased PMN-PT single crystal/Terfenol-D ME sensor.
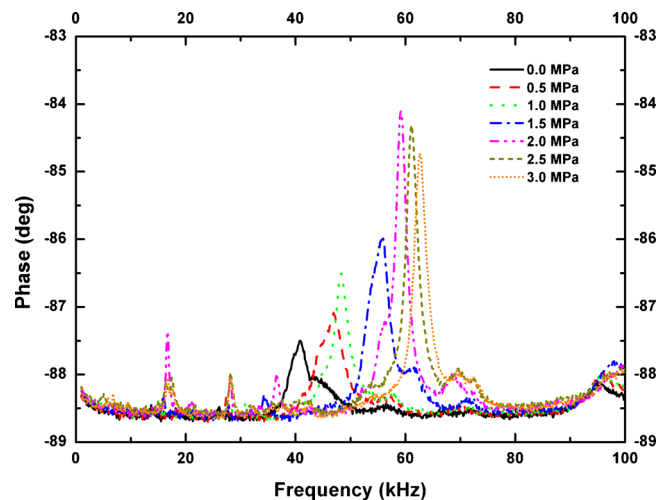

FIG. 3. (Color online) Phase vs frequency spectrum of a PMN-PT single crystal/Terfenol-D ME sensor under different stress-biased conditions.

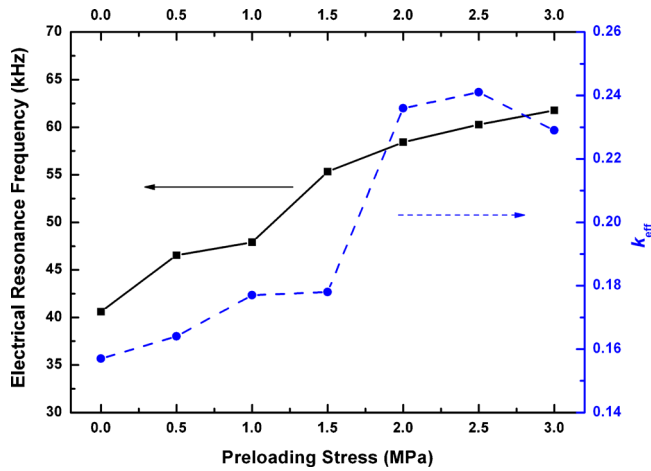FIG. 4. (Color online) Electrical resonant frequency and k_{eff} of a PMN-PT single crystal/Terfenol-D ME sensor obtained in an impedance measurement as a function of preloading stress.

$$\frac{\Delta f}{f_o} = \frac{f - f_o}{f_o} = AF, \tag{2}$$

where f_o is the fundamental resonant frequency of the sensor under test, f the resonant frequency under pressure/force, Δf the change in resonant frequency, A a constant, and F is the applied force. Since the resonant frequency of the piezoelectric material is inversely proportional to its thickness, the negative sign implies that the resonant frequency increases when the force/pressure is applied on the piezoelectric material.

Besides the increase in the resonant frequency, the resonance mode becomes stronger when the preloading stress increases. With the technique of mechanical preloading, the PMN-PT single crystal can function in both contraction and expansion. Under a specific stress, the energy coupling between the PMN-PT single crystal and Terfenol-D alloy is enhanced. To further evaluate the electrical performance of the ME sensor, the effective electromechanical coupling coefficient, k_{eff}, is determined by the following:[18]

$$k_{\text{eff}} = \sqrt{\frac{f_p^2 - f_s^2}{f_p^2}}, \tag{3}$$

where f_s and f_p are the series and parallel resonant frequencies of the fundamental mode of the transducer, respectively. This coefficient describes the conversion of energy from electrical to mechanical form or vice versa. Figure 4 shows the dependences of the resonant frequency and k_{eff} of the ME sensor under the effect of different preloading stress. The resonant frequency of the ME sensor increases monotonically with the preloading stress. On the other hand, the k_{eff} coefficient increases gently at low preloading stress and reaches a peak at 2.5 MPa. Since the single crystal cracks or even breaks into pieces under further increase in preloading stress, the k_{eff} drops gradually after reaching a peak value.

Figures 5 show the frequency dependence of the ME voltage coefficient of the sensor under different stress-biased conditions. Figure 5(b) shows the ME voltage coefficient of the sensor in the low frequency region (below 15 kHz). When the sensor is used to measure an unknown ac magnetic
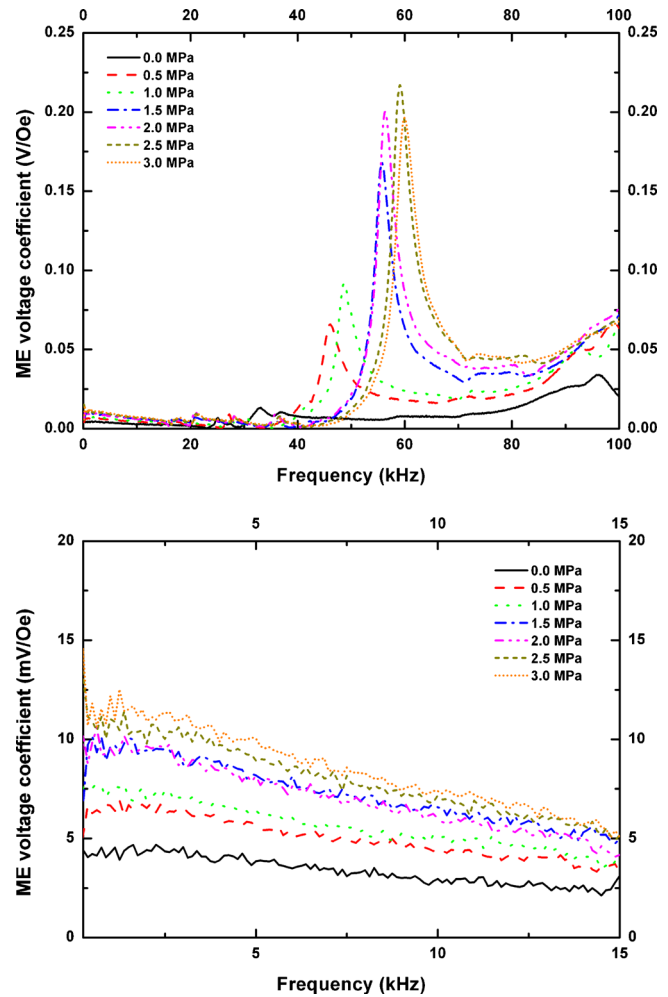

FIG. 5. (Color online) Frequency dependence of ME voltage coefficient as a function of preloading stress for a PMN-PT single crystal/Terfenol-D ME sensor. (Inset: a magnified picture of low frequency region.)

field, from the measured voltage and known ME coefficient at that frequency, strength of the magnetic field can be estimated. The ME resonance peak shifts to higher frequency as the preloading stress increases. The pattern shown in Fig. 6 is
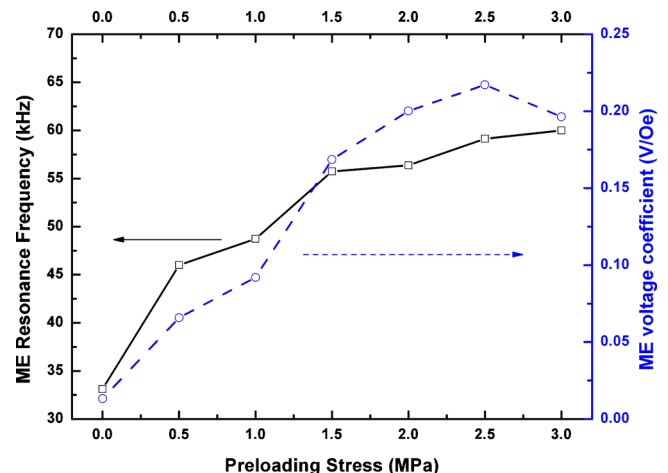

FIG. 6. (Color online) ME resonant frequency and ME voltage coefficient of a PMN-PT single crystal/Terfenol-D ME sensor obtained in an electrical impedance measurement as a function of preloading stress.

similar to that obtained in the electrical impedance measurements (Fig. 4). At frequencies away from the resonant frequency, the ME voltage coefficient of the sensor increases with increasing preloading stress. Compared to the free condition, the ME performance of the sensor is enhanced significantly under specific stress-biased conditions.

We believe that the main contribution to the enhanced ME affect comes from non-180° domain switching effect in the piezoelectric PMN-PT crystal and a mechanical coupling enhancement between the piezoelectric and magnetostrictive materials. In the piezoelectric PMN-PT single crystal, a certain level of the preloading stress would enhance the possibility of domain switching and the generation of non-180° domains. The switching of the non-180° domains induced by the vibration of Terfenol-D rod results in enhanced piezoelectric performance. While in the Terfenol-D rod, the preloading stress along its longitudinal-axis direction causes the magnetization vector of the magnetic domains to change from its random orientation to the direction perpendicular to the longitudinal-axis. When H_{ac} is applied along the longitudinal-axis direction, maximum magnetostriction can be achieved. With the stress-biased enhancement of piezoelectricity and magnetostriction as well as the improved electromechanical coupling, higher ME voltage coefficient can be obtained. In the sensor described, the ME performance of the sensor increases with increasing preload even in the low frequency region (<15 kHz). At resonance, the maximum ME voltage coefficient of the ME sensor approaches 0.22 V/Oe under a preload of 2.5 MPa. After that saturation point, microcracks began to be induced on the crystal, which would degrade the performance of the sensor. Besides, when the preloading stress exceeds a critical compressive stress, non-180° domains of the crystal are going to be removed gradually.[7] We believe that 2.5 MPa may be the critical compressive stress of the sensor. Therefore, both the piezoelectric and magnetostrictive effects will be reduced with further increased preload beyond the critical stress.

IV. CONCLUSION

A stress-biased magnetoelectric sensor using piezoelectric PMN-PT single crystal and magnetostrictive Terfenol-D alloy composite has been fabricated. A built-in permanent magnet installed in a metal frame is used to provide a dc magnetic bias on the PMN-PT/Terfenol-D ME sensor. The resonance characteristics and magnetoelectric performance of the sensor have been evaluated under different stress-biased conditions. With increasing preloading stress, the resonance of the sensor shifts to higher frequency. At a preloading stress of 2.5 MPa, the sensor is found to exhibit a giant ME voltage coefficient of 0.22 V/Oe at resonance. This is due to the enhanced piezoelectric and magnetostrictive effects, as well as the enhanced mechanical coupling between these two materials under the preloading stress. With a built-in permanent magnet, this high ME-performance sensor has the potential in practical uses such as magnetic-field sensing or magnetic energy converting applications.

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