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Ultralow equivalent magnetic noise in a magnetoelectric Metglas/Mn-doped $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ heterostructure

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An ultralow equivalent magnetic noise of 6.2 pT/$\sqrt{\text{Hz}}$ at 1 Hz was obtained in a bimorph heterostructure sensor unit consisting of longitudinal-magnetized Metglas layers and a transverse-poled 1 mol. % Mn-doped $Pb(Mg_{1/3}Nb_{2/3})O_3$-$29PbTiO_3$ (PMN-PT) single crystal. Furthermore, the equivalent magnetic noise was ≤ 1 pT/$\sqrt{\text{Hz}}$ at 10 Hz. Compared with previously reported multi-push-pull configuration Metglas/PMN-PT sensor units, the current heterostructure exhibits a higher magnetoelectric coefficient of 61.5 V/(cm × Oe), a similar equivalent magnetic noise at 1 Hz and a lower noise floor at several hertz range. The ultralow equivalent magnetic noise in this sensor unit is due to the low tangent loss and ultrahigh piezoelectric properties of Mn-doped PMN-PT single crystals. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4733963]

Multiferroic magnetoelectric (ME) materials represent an appealing class of multifunctional materials that simultaneously exhibit electric and magnetic orderings. The coexistence of two order parameters brings about physical phenomena and offers potential for devices.[1,2] The ME effect can be realized as intrinsic in single-phase compounds and as extrinsic in strain-mediated multi-phase composites. Since no single-phase material has been found that demonstrates a practical capacity for such coupling at room temperature, multi-phase composites consisting of ferroelectric and ferromagnetic phases have drawn significant interest in recent years due to the potential application in passive magnetic field sensors.

An important and basic challenge remaining to be fulfilled for practical technological use of ME composites is the realization of high magnetic field sensitivity, which is determined not only by the output signal of the composites in response to an incident magnetic field but also by the equivalent magnetic noise generated in the absence of an incident field. To tackle this challenge, two corresponding strategies have been employed: (1) enhancement of ME coefficients and (2) reduction of equivalent magnetic noises. For example, composite configurations with high effective energy transduction, component phases with large individual properties,[3] and interface optimization with strain-engineering[4,5] have been studied to enhance the ME coefficients and magnetic field sensitivity. Alternatively, techniques to reduce the equivalent magnetic noise in ME composites need to focus on dominate intrinsic noise sources of the sensor, namely dielectric loss tanδ (i.e., $N_{DE} = \sqrt{\frac{4kTC_P \tan\delta}{2\pi f}}$) and dc leakage resistance R_{dc} (i.e., $N_R = \frac{1}{2\pi f}\sqrt{\frac{4kT}{R_{dc}}}$). Accordingly, the total noise charge density (N_t) is given by Ref. 3.

$$N_t = \sqrt{N_{DE}^2 + N_R^2} = \sqrt{\frac{4kTC\tan\delta}{2\pi f} + \frac{1}{(2\pi f)^2}\frac{4kT}{R_{dc}}}, \qquad (1)$$

where k is Boltzmann's constant (1.38×10^{-23} J K^{-1}), T is the temperature in Kelvin, C is the capacitance of the sensor, and f is the frequency in Hertz. Clearly, the reduction in equivalent magnetic noise focuses on the decrease of tanδ and increase of R_{dc}.

Single crystal ferroelectrics, such as lead magnesium niobate-lead titanate (PMN-PT) near the morphtropic phase boundary between ferroelectric rhombohedral and ferroelectric tetragonal phases, exhibit ultrahigh piezoelectric coefficients of ~2000 pC/N and low tangent losses of ~0.5%: as listed in Table I. In particular, it has been found that Mn substitutions in PMN-PT are effective in achieving higher coercive fields, and lower dielectric permittivities and tangent losses due to the selectively pining of 180° domain wall motions.[6] For example, in ⟨011⟩-oriented 1 mol. % Mn-doped PMN-29PT, giant piezoelectric coefficient $d_{31} = -1800$ pC/N and extremely low tan$\delta = 0.07\%$, have been reported,[6] as summarized in Table I. The superior properties of Mn-doped PMN-PT single crystal provide opportunities for realization higher magnetic field sensitivity through a combination of giant ME effects and ultralow equivalent magnetic noise.

In this letter, we report a longitudinal-magnetized Metglas and transverse-poled Mn-doped PMN-PT bimorph heterostructure, which simultaneously exhibits a giant ME coefficient of 61.5 V/(cm × Oe) and a low equivalent magnetic noise of 6.2 pT/$\sqrt{\text{Hz}}$. Compared with multi-push-pull mode Metglas/piezofiber sensors, this L-T mode Metglas/Mn-doped PMN-PT bimorph heterostructure had not only higher ME coefficients and close equivalent magnetic noise floors but also had additional advantages, including: properties repeatability, (i.e., reduction of tanδ contribution from interfacial epoxy layer[4]), fabrication flexibility, low cost, and device miniaturization.

Figure 1 shows a schematic diagram and a photo of the ME Metglas/Mn-doped PMN-PT bimorph heterostructure.

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TABLE I. Property parameters for multi-push-pull mode Metglas/PMN-PT and L-T mode Metglas/Mn-doped PMN-PT sensors and the related pure single crystals.

ε_{33}	tanδ	d_{33} or d_{31} [pC/N]	C [pC]	tanδ	R_{dc} [GΩ]	α_E [V/(cm × Oe)]	α_Q [pC/Oe]
⟨001⟩-PMN-PT[a]			M-P-P mode Metglas/PMN-PT[c]				
7000	0.005	2000	344	0.008	80	52	2680
<110>-Mn-doped PMN-PT[b]			L-T mode Metglas/Mn-doped PMN-PT				
1300	0.001	1800	3120	0.0014	10	61.5	3914

[a] Cited from Ceracomp Co., Ltd.

[b] Measured based on IEEE standards.

[c] Cited from Ref. 3.

High-quality Mn-doped PMN-PT single crystals were grown directly from a melt by a modified Bridgman technique.[7] As-grown single crystals were oriented along ⟨001⟩, ⟨011⟩, and ⟨0-11⟩ directions, diced to prepare fibers of dimensions of 30 × 2 × 0.2 mm^3, and the fibers arranged with their ⟨001⟩ and ⟨011⟩ crystallographic axes oriented in the length and thickness directions. It has been shown in our previous work that PMN-PT plates, specially cut and poled along the ⟨011⟩ thickness direction, possess higher transverse piezoelectric coefficients (i.e., an ultrahigh thickness direction voltage response to a length direction strain deformation).[8] After deposition of gold electrodes on the thickness surfaces, the Mn-doped PMN-PT crystal fibers were poled under an electric field of 1500 V/mm (5 times of coerive field) at 120 °C for 15 min in sillicon oil and 750 V/mm on cooling thereafter. Metglas were commercially supplied with composition $Fe_{74.4}Co_{21.6}Si_{0.5}B_{3.3}Mn_{0.1}C_{0.1}$ (Vacuumscheltze GmbH & Co. KG, Germany) as a roll with a thickness of 25 μm, and were cut into foils of dimensions 80 × 8 mm^2. Twelve such Metglas layers were then stacked one on top of each other, and bonded with epoxy resin (West system 206, USA) using a vacuum bag pressure method. The poled Mn-doped PMN-PT plate was then stacked and bonded to the center of Metglas layers using a nonconductive epoxy resin (Westsystem, USA) and a spot of Silver paint (Ted Pella, Inc.) at its ends.

Measurement of the ME voltage coefficient α_E for Metglas/Mn-doped PMN-PT bimophor heterostructures was done by a derivative measurement determined from the ME charge coefficient α_Q, capacitance C of the sensor, and the thickness of the piezoelectric layer (i.e., $\alpha_E = {}^{\alpha_Q}\!/_{C \times t}$). Thus, the dielectric properties of the heterostructure were first measured using an impedance analyzer (Agilent 4294 A). The dc resistance R_{dc} was determined to be 10 GΩ using a pA Meter/DC voltage source (HP 4140 B) based on Ohm law. The parameters of the heterostructure are listed in Table I.

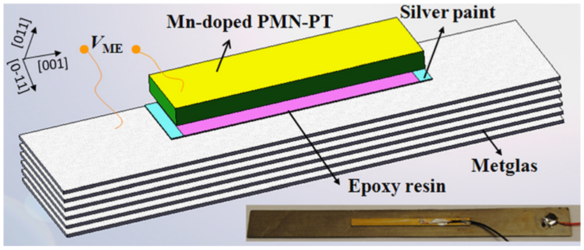

FIG. 1. Schematic diagram and photograph of the proposed Metglas/Mn-doped PMN-PT bimorph heterostructure.

The ME charge coefficient α_Q was measured by a charge meter (Kistler type 5015), coupled with a lock-in amplifier (Stanford Research, SR-850), as a function of dc magnetic bias field H_{dc} in response to a constant ac magnetic drive of $H_{ac} = 0.1$ Oe at frequency $f = 1$ kHz for various Metglas layers N (where N was varied by successively peeling off layers). Both the excitation magnetic field and dc bias were applied along the length of the heterostructure. The measured results were then converted to α_E, as shown in Fig. 2(a). From this figure, it can be seen that the values of α_E for laminate with different N were nearly zero at $H_{dc} = 0$; dramatically increased as H_{dc} was increased; reached a maximum at a particular H_{dc}; and subsequently decreased as H_{dc} was further increased. Figure 2(b) shows a summary of the data given in Fig. 2(a), and reveals that the variation of the maximum value of α_E with N for the bimorph heterostructure. The value of α_E did not dramatically decrease after reaching a maximum with increasing N, as previously reported in sandwiched laminate.[9] A high value of $\alpha_E = 61.5$ V/(cm × Oe) can be seen at $N = 5$, which was 1.2 × enhancement relative to $N = 6$. It should be noted that prior reports for laminated composites have focused on $N = 6$.[3,5,10] An abnormal thickness

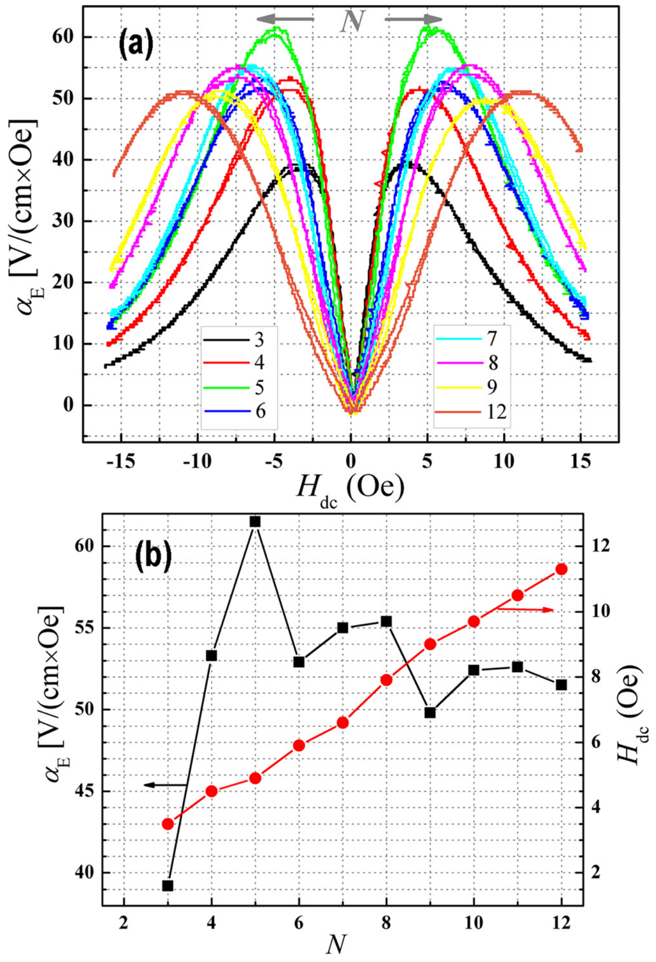

FIG. 2. (a) ME coefficient α_E as a function of dc magnetic bias field H_{dc} for various layers of Metglas. The numbers indicate the number of Metglas layers (N). (b) Maximum α_E and the required optimal H_{dc} dependence of the number of Metglas of Metglas layers N.

fraction-dependent α_E for bending mode ME laminates has been previously discussed elsewhere.[11]

We next measured the induced charge of the bimorph heterostructure for $N = 5$ (optimal number of Metglas layers) as a function of f under $H_{ac} = 0.05$ Oe and $H_{dc} = 5$ Oe over the range of $1\,\text{Hz} < f < 50\,\text{kHz}$. Figure 3(a) shows a strong enhancement of α_Q to $\sim$80 nC/Oe at $f = 25$ kHz due to the longitudinal electromechanic resonance (EMR). Figure 3(b) presents a partially enlarged view for $1\,\text{Hz} < f < 1\,\text{kHz}$. In addition to the enhancement due to the longitudinal EMR, three additional weaker peaks were observed from bending mode resonance caused by an asymmetrical stress distribution in the bimorph structure. Correspondingly, three anti-resonance peaks were also observed, where α_Q exhibited a dip in value. These behaviors of various bending mode laminates have previously been reproted.[10,11] In order to experimentally indentify the enhancement in α_Q, the electrical impedance spectrum was measured over the range of $70\,\text{Hz} < f < 1\,\text{kHz}$ using an Agilent 4294 impedance analyzer. It was found that the value of α_Q was maximized at the resonance frequencies. In Figure 2(b), it can also be seen that α_Q was relatively insensitive to frequency for $1\,\text{Hz} < f < 70\,\text{Hz}$: i.e., 4087 pC/Oe at 1 Hz, and 3948 pC/Oe at 70 Hz.

More recently, we have reported an extremely low equivalent magnetic noise in a multi-pull-pull (M-P-P) mode Metglas/PMN-PT sensor.[3] Here, the noise charge density and the equivalent magnetic noise of the current L-T mode Metglas/Mn-doped PMN-PT and the previously reported M-P-P Metglas/PMN-PT sensors are compared, as shown in Fig. 4. First, the noise charge density due to tanδ and R_{dc} of the two sensors presented in Fig. 4(a) were estimated using appropriate sensor parameters (summarized in Table I). For the M-P-P mode sensor, both tanδ and R_{dc} noises contributed to the total noise charge density at 1 Hz, but the magnitude of the tanδ noise was 1.2 × larger than that of the R_{dc} noise. However, for the L-T mode sensor, the total noise charge density at 1 Hz was dominated by R_{dc} noise, and the magnitude of R_{dc} noise was 45.5 × larger than that of the tanδ noise: this is because tanδ of the L-T mode sensor was extremely low (tan$\delta = 0.0014$) and R_{dc} was relatively low. It can be seen that the predicted 1 Hz noise charge density of the L-T mode sensor was 2 × higher than that of M-P-P mode one. This is because of the much higher R_{dc} noise of the L-T mode sensor, even though the tanδ noises of the two modes were close. In this figure, the measured noise charge density of the L-T mode Metglas/Mn-doped PMN-PT sensor is also given. Except at frequencies where external vibration sources were present, the estimated and measured noise

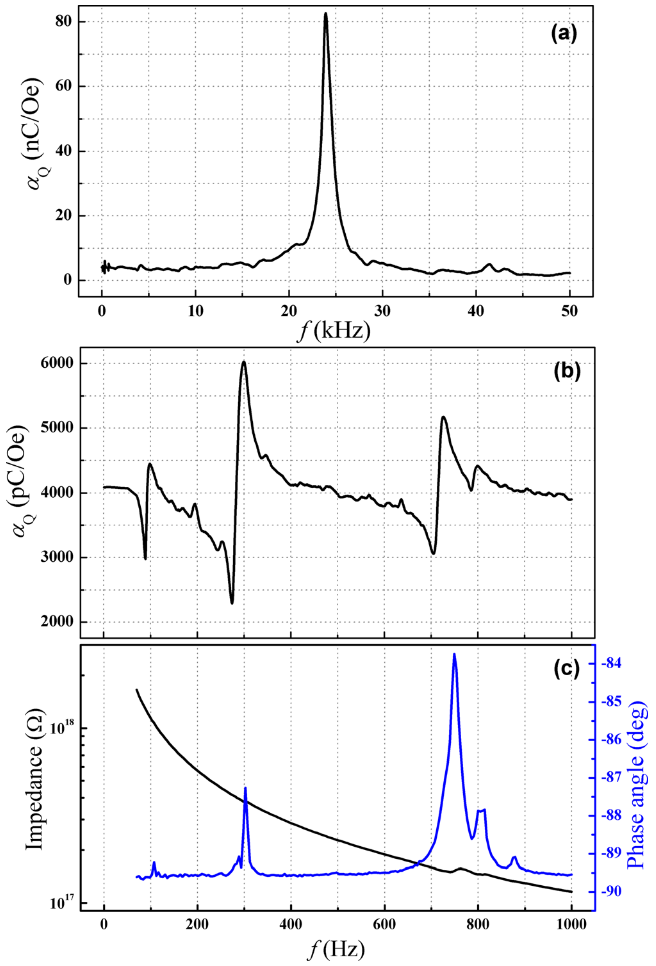

FIG. 3. (a) ME charge coefficient α_Q as a function of frequency f and (b) the close-view for $1\,\text{Hz} < f < 1\,\text{kHz}$. (c) Impedance spectrum of the heterostructure over the frequency rage $70\,\text{Hz} < f < 1\,\text{kHz}$.

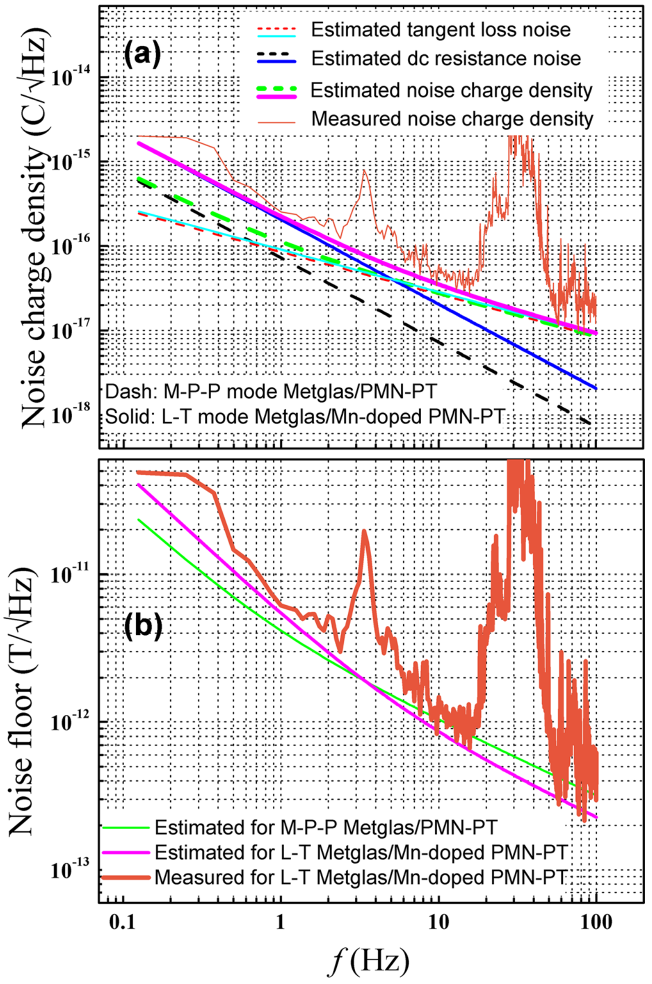

FIG. 4. (a) Measured and estimated noise charge density and (b) equivalent magnetic noise for the multi-push-pull mode Metglas/PMN-PT sensor unit (Ref. 3) and the proposed one over the frequency range of 0.125 Hz $< f <$ 100 Hz. The noise contributions, including constituent dielectric loss and dc resistance loss, are compared.

charge density exhibited good agreement, especially for 1 Hz and 10 Hz.

The estimated and measured equivalent magnetic noise spectra given in Fig. 4(b) were obtained through a conversion of the noise charge density spectrum [see Fig. 4(a)] and the ME charge coefficient [see Fig. 3(b)]. Please note that the value of α_Q used here was 4087 pC/Oe, which was nearly constant for $1\,\text{Hz} < f < 70\,\text{Hz}$. Even though the value of α_Q varied for $70\,\text{Hz} < f < 100\,\text{Hz}$, it was not necessary to use a mutative value of α_Q: as the noise charge density was dominated by external vibration noise sources. The results show that the estimated 1 Hz equivalent magnetic noise of the L-T mode was still 1.3 × times higher than the M-P-P mode one (i.e., 5.5 pT/$\sqrt{}$Hz to 4.2 pT/$\sqrt{}$Hz), whereas the 10 Hz value was 1.3 × times lower (i.e., 0.8 pT/$\sqrt{}$Hz relative to 1.0 pT/$\sqrt{}$Hz). These results show that our L-T mode Metglas/Mn-doped PMN-PT sensor has a 1 Hz equivalent magnetic noise close to that previously reported for M-P-P mode Metglas/PMN-PT sensors, with a noise floor that drops off more rapidly with increasing frequencies. Experimentally, an ultralow equivalent magnetic noise of 6.2 pT$\sqrt{}$Hz was found at 1 Hz, which was close to the predicted value of 5.5 pT$\sqrt{}$Hz. Please note that the equivalent magnetic noise of the proposed ME sensor unit was lower than 1 pT$\sqrt{}$Hz at $f = 10\,\text{Hz}$. Furthermore, the current L-T mode Metglas/Mn-doped PMN-PT laminate has much lower equivalent magnetic noise at interesting $f = 1\,\text{Hz}$ than the similar L-T mode Metglas/pure PMN-PT laminate (i.e., 6.2 pT/$\sqrt{}$Hz to 10.8 pT/$\sqrt{}$Hz).[12] These low equivalent magnetic noises observed for the L-T mode originate from a low tanδ noise, coupled with a giant ME charge coefficient.

In summary, an ultralow equivalent magnetic noise has been found in a L-T mode Metglas/Mn-doped PMN-PT bimorph heterostructure, coupled with a low noise charge amplifier. In particular, the measured equivalent magnetic noise for this sensor unit was 6.2 pT$\sqrt{}$Hz at 1 Hz, which decreased to ≤ 1 pT$\sqrt{}$Hz at $f = 10\,\text{Hz}$. Analysis has shown that this ultralow equivalent magnetic noise is due to a low tangent loss and a giant ME coefficient. These results indicate that Metglas/Mn-doped PMN-PT heterostructures are a good candidate for realization of ultralow magnetic field detection applications.

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