

Magnetoelectric Sensors

Subjects: [Materials Science](#), [Ceramics](#)

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Multiferroic magnetoelectric (ME) materials with the capability of coupling magnetization and electric polarization have been providing diverse routes towards functional devices and thus attracting ever-increasing attention. The typical device applications include sensors, energy harvesters, magnetoelectric random access memories, tunable microwave devices and ME antennas etc. Among those application scenarios, ME sensors are specifically focused in this review article. We begin with an introduction of materials development and then recent advances in ME sensors are overviewed. Engineering applications of ME sensors are followed and typical scenarios are presented. Finally, several remaining challenges and future directions from the perspective of sensor designs and real applications are included.

multiferroic

magnetoelectric

sensors

object detection

magnetic localization

current sensing

biological magnetic measurement

non-destructive testing

displacement sensing

1. Introduction

Multiferroic materials have been recently attracting ever-increasing attention because of the capability of coupling at least two ferric orders, i.e., ferroelectricity, ferromagnetism, or ferroelasticity, and the vast potential for multifunctional devices applications ^{[1][2][3][4][5]}. A control of polarization P by external magnetic field H (direct ME (DME) effect) or a manipulation of magnetization M by an electric field E (converse ME (CME) effect) can be realized in multiferroic magnetoelectric (ME) materials ^[6]. Compared with single-phase ME material, ME heterostructures and ME laminates perform greatly enhanced coupling capability, which is generally characterized by ME coefficient α_{ME} ^{[7][8][9]}. After a development of nearly half a century, tremendous progress regarding ME composites and related device applications has been reported ^{[1][2][3][6][10][11][12][13][14][15][16][17][18][19]}.

2. Materials for ME Sensors

The ME effect was first experimentally demonstrated in single-phase multiferroic material Cr_2O_3 in 1961 ^{[20][21]}. After that, diverse studies all over the globe were conducted to further enhance the coupling capability of ferroelectric and magnetic orderings in a single-phase material system ^{[20][22]}, but the low Curie temperature and the weak ME coupling capability in single-phase ME materials, such as BiFeO_3 , BiMnO_3 and LuFe_2O_4 , greatly limited their applications ^{[1][23][24]}. The proposal of a product effect in composite ME materials by combining the

piezomagnetic and piezoelectric effects of ferromagnetic and ferroelectric materials then provided new routes towards improved ME coupling performance. Early in 1986, Pantinakis et al. proposed 2-2 type ME composites based on the aforementioned product effect [25] and giant ME coefficients were gradually realized in laminated ME composites starting from the beginning of 21st century [1][6][10]. Compared with single-phase or 0-3 typed ME materials, 2-2 typed ME composites, such as a bulk ME laminates with piezoelectric phase ($\text{Pb}(\text{Zr,Ti})\text{O}_3$ (PZT), $\text{Pb}(\text{Mg,Nb})\text{O}_3$ - PbTiO_3 (PMN-PT)) embedded in piezomagnetic materials (FeCoSiB, FeBSiC Terfenol-D, Ni or Fe-Ga) [8] and a FeGaB/AlN thin-film ME heterostructure [26], exhibited enhanced ME coupling performance benefitting from the removal of the leakage current and the improvement of the interfacial strain transfer. At this section, we will first review materials advances in ME sensors since 2002.

2.1. Bulk ME Laminates

It is highly desirable to design new connectivity structures for circumventing the limitation of leakage current that occurs in 0-3 typed ME composites. Back in 2002, Ryu et al. developed a laminated Terfenol-D/PZT/Terfenol-D ME composite (Figure 1a) with 2-2 type connectivity to solve the leakage current problem in 0-3 type ME composites, and the obtained ME coupling coefficient at non-resonance frequency reached as high as 5 V/cm·Oe [27]. This was a significant event in the development of ME laminates and various kinds of laminated structures were proposed afterwards [10][27]. For example, Dong et al. reported 2-2 type ME laminates consisting of Terfenol-D ferrite and PMN-PT piezoelectric crystal. These ME composites work with L-T mode and display relatively low ME coefficients of 2.2 V/cm·Oe at non-resonance frequency [28]. In a bid to further improve the ME voltage coefficient, Dong et al. in 2005 first proposed a push-pull mode that increased the distance between electrodes and decreased the static capacitance of ME laminates from nF to pF scale [29][30]. In such 2-2 type ME composites, the piezoelectric core was symmetrically poled along its longitudinal direction and rgw d_{33} piezoelectric constant of a piezoelectric material could be utilized. A giant ME voltage coefficient of 1.6 V/Oe at non-resonant frequencies was observed experimentally [30]. One year later, Dong et al. further developed a multi-push-pull mode in 2-1 ME composites. The schematic structure configuration and operation mode of such a 2-1 ME composite is presented in Figure 1c. It consisted of a piezo-fiber layer laminated between FeBSiC alloys. For the first time, the non-resonant ME coefficient at 1 Hz reached 22 V/cm·Oe, making such a structure especially suitable for low-frequency and passive magnetic sensing [31][32][33][34][35], but it should be noted here that the mechanical quality factor for such a 2-1 type ME composites is normally less than 100, so ultra-high resonant ME coefficients cannot be realized in this case [29].

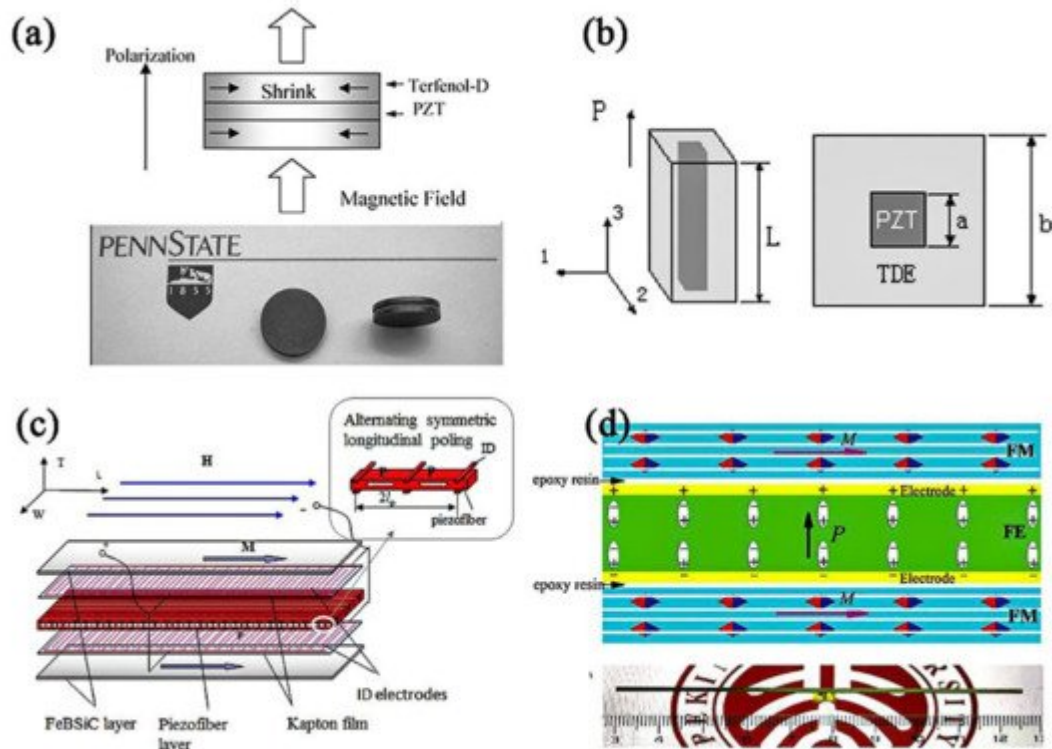


Figure 1. (a) Schematic structure (top) and photograph (bottom) of ME laminate composites using Terfenol-D and PZT disks [27]. (b) 3D and cross sectional schematic illustration of the single period of 1-3-type ME structure [36]. (c) Illustration of the FeBSiC/piezofiber laminate configuration working on multi-push-pull mode [29][30]. (d) The schematic view for 1-1 laminated ME composite and a(ii) the prototype snapshot of the 1-1 typed ME sample [8].

Another way to address the difficulty of fully polarizing the piezoelectric phase in 0-3 type ME composites is replacing the particle phase with a 1-D piezoelectric fiber (forming 1-3 typed connectivity). For example, in 2005 Nan et al. reported a 1-3 type ME composite with ZT rod arrays embedded in a Terfenol-D medium via a dice-and-fill technique. The non-resonant ME coupling coefficient reached $6.2 \text{ V/cm}\cdot\text{Oe}$ [37], which represented great progress for ME composites. Two years later, Ma et al. simplified this 1-3 type ME structure by just embedding one single PZT rod in a Terfenol-D/epoxy mixture [36]. The single period element of the 1-3 ME composites is shown in Figure 1b. Although the non-resonant ME coupling coefficient decreased by almost one order of amplitude, this simple structure, low-cost fabrication process and sub-millimeter size made it attractive for micro-ME array applications [36].

In 2017, Chu et al. reported a 1-1 type ME composites, which consisted of a [011]-oriented $\text{Pb}(\text{Mg},\text{Nb})\text{O}_3\text{-PbZrO}_3\text{-PbTiO}_3$ (PMN-PZT) single crystal fiber and laser-treated amorphous alloy Metglas. The 1-1 type ME composite featured the one-dimensional configuration as shown in Figure 1d [8]. The laser treatment could decrease magnetic hysteresis loss of Metglas and thereby enhance the Q value of the ME resonator. In addition, the fiber configuration effectively utilized the magnetic flux concentration effect occurring in Metglas layers. More importantly, this 1-D configuration favored the longitudinal vibration mode of ME laminates. A ME coupling coefficient of $\sim 7000 \text{ V/cm}\cdot\text{Oe}$, that was nearly seven times higher than the best result published previously, was finally realized,

opening a door to develop new ME devices, e.g., resonant magnetic receivers in particular [8]. In addition, a high ME coefficient of 29.3 V/cm·Oe at non-resonant frequency was also achieved for our 1-1 type composites. Note, only one single crystal was consumed in this case, while previous 2-1 type composites normally took five crystals. In 2020, the resonant ME coefficient of 1-1 type ME composites was further enhanced to 12,500 V/cm·Oe by using a hard piezo-crystal Mn-PMN-PZT [9]. A summary of the field coupling coefficient of different ME laminates, i.e., 0-3, 2-2, 2-2.1-1 ME laminates, is given in Table 1.

Table 1. Some ME laminates and their ME coupling performances.

Composition	Year	Connectivity	Working Mode	$\alpha_{ME}^{non-resonance}$ (V/cm · Oe)	$\alpha_{ME}^{resonance}$ (V/cm · Oe)
Terfenol-D/PZT [36]	2007	3-1	L-L	0.5	18.2
NiFe ₂ O ₄ /PZT [38]	2001	2-2	L-T	1.5	/
Terfenol-D/PZT [27]	2002	2-2	L-T	5	/
Metglas/PVDF [39]	2006	2-2	L-T	7.2	310
Metglas/P(VDF-TrFE) [40]	2011	2-2	L-L	17.7	383
Lanthanum gallium tantalite/permendur [41]	2012	2-2	/	2.3	720
FeCoSiB/(Pt)/AlN in vacuum [42]	2013	2-2	L-T	/	20,000
FeCoSiB/(Pt)/AlN [43]	2016	2-2	L-T	/	5000
Metglas/LiNbO ₃ [44]	2018	2-2	L-T	1.9	1704

References

FeBSiC/PZT [30]	2006	2-1	L-L	22	500		es: From
Metglas/PMN-PT [31]	2011	2-1	L-L	45	1100		novel
Metglas/PMN-PT without laser treatment [8]	2017	1-1	L-T	29.3	5500		electric res. NPJ
Metglas/PMN-PT with laser treatment [8]	2017	1-1	L-T	22.9	7000		ys. D
Metglas/Mn-PMN-PZT with laser treatment [9]	2020	1-1	L-T	23.6	12,500		l. Phys.
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9. PourhosseiniAsl, M.; Gao, X.; Kamalisiahroudi, S.; Yu, Z.; Chu, Z.; Yang, J.; Lee, H.-Y.; Dong, S. Versatile power and energy conversion of magnetoelectric composite materials with high efficiency via electromechanical resonance. *Nano Energy* 2020, 70, 104506. Note: Connectivity. We use different numbers to represent the connectivity of each individual phase. For example, 1-3 type composite means one-phase fiber (denoted by 1) was embedded in the matrix of another phase (denoted by 3); 2-2 type composite means laminated structure (each phase has a plane configuration denoted by 2); 2-1 type composite means one-phase fiber was laminated with another phase plate; 1-1 type means both phases are in the form of fiber configuration. *Working mode*. L-L, L-T means longitudinal vibrations with longitudinal magnetization and transverse polarization (L-L) or transverse magnetization and transverse polarization (L-T).

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2.2. MEMS and NEMS ME Laminates

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- Figure 2.** Sketch of ME MEMS cantilever with the functional layer deposited on one side (a) [43] and two side (b) [50]. (c) and (d) show the suspended circular plate and AlN anchors. The yellow area presents the electrode [53].
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3. Advances in ME Sensors

The giant ME coupling in ME composites provides the chances to be implemented as diverse functional devices, such as sensors, energy harvesters, magnetoelectric random access memories, tunable microwave devices and

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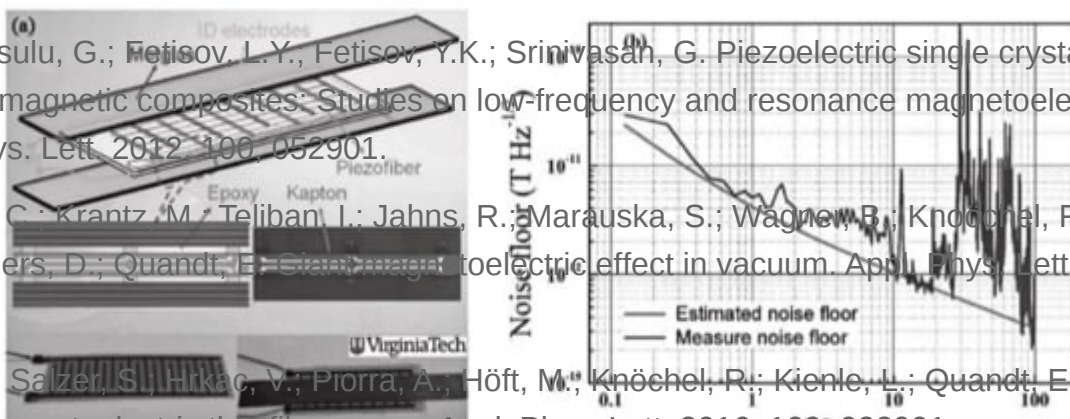


Figure 4. (a) 3D structure of Metglas/Mn-PMNT ME composite for sensing of magnetic fields. Appl. Phys. Lett. 2018, 112, 262206. (b) Measured and estimated equivalent magnetic noise of the proposed sensor unit [33].

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Figure 4. 3D structure of Metglas/Mn-PMNT ME composite (a) and its cross-sectional diagram (b), (c) The EMN over the frequency range of 8 Hz < f < 100 Hz. (d) The EMN and Nt of different Metglas/Mn-PMNT sensors at 30 Hz [55].

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observed by a mixed signal; Refs. [56]. The base sensor was a high-shield frequency conversion in the modulation frequency to electric composite for quasi-static magnetic field detection. Appl. Phys. Lett. 2013, 103, 211202.

amplitude modulation (envelope) signal was generated due to the intrinsic frequency mixing characteristic in ME sensors, as shown in Figure 5a(ii).

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The measured output waveform in response to an applied weak AC magnetic field at 100 mHz. (d) A linear-response to varying H_{AC} at 100 mHz with a step of 0.1 nT [55].

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In order to test the limit of detection by using this amplitude modulation method, the time constant decreased to 10 ms and the demodulated signal from time domain waveform via a lock-in amplifier was analyzed. Figure 5c shows

64. Chu, Z.; Shi, H.; Pourhosseini-Asl, M.; Wu, J.; Shi, W.; Gale, X.; Yuan, X.; Dong, S. A linear-response to AC with this range was obtained as given in the inset in Figure 5c. Accordingly, the limit of resolution (LOR) of the ME sensor based on this amplitude modulation method was determined to be as low as

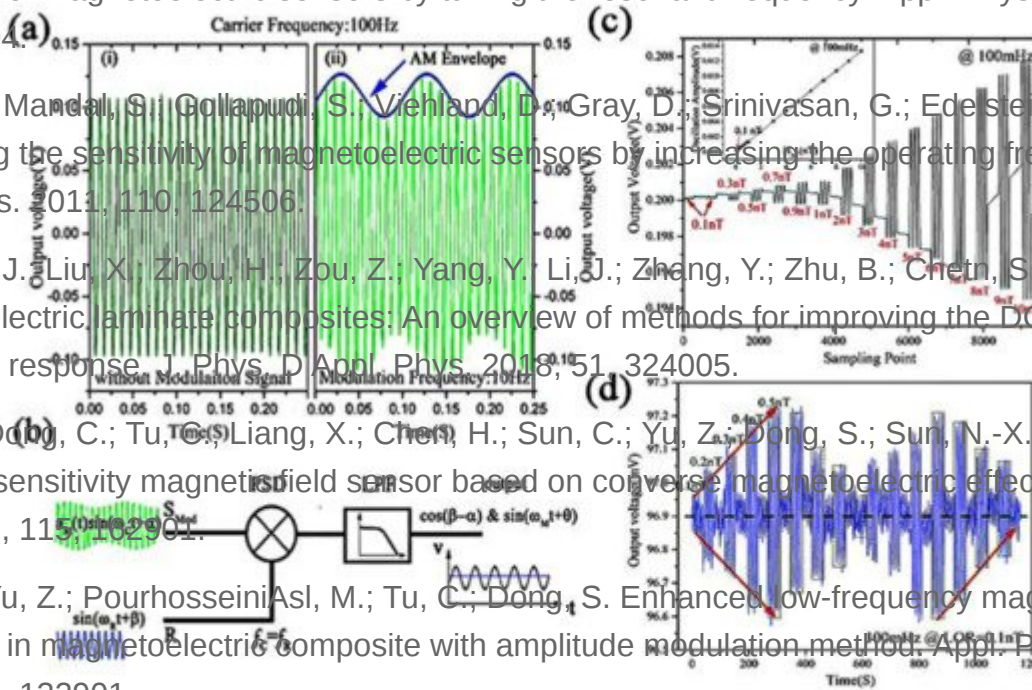
100 pT. To confirm this LOR, Figure 5d further verified it by measurement. Considering an equivalent noise bandwidth (ENBW) of 7.8 Hz corresponding to the given measurement system, the calculated LoD was then calculated as 33 pT/√Hz at 0.1 Hz.

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sensing based on the nonlinear magnetoelectric effect in magnetic heterostructures. J. Phys. D Appl. Phys. 2016, 49, 375002.

ME laminates can be viewed as resonators from the perspective of mechanics and resonant phenomenon is also able to enhance the ME coupling coefficient and thus to improve the detection ability [10]. In this regard, ME

67. Burdin, D.A.; Ekonomov, N.A.; Chashin, D.V.; Fetisov, L.Y.; Fetisov, Y.K.; Shamoin, M. Temperature Dependence of the Resonant Magnetoelectric Effect in Layered Heterostructures.



Materials 2017, 10, 2118. In a 1-1 type ME composite, Dong et al. reported an enhanced LoR of 1.2 pT early in 2005 (see Figure 6a) [29]. As for MEMS ME magnetic sensor, Yarar et al. developed a low temperature deposition route of very high quality AlN film, allowing the reversal process flow. Correspondingly, the LoD was enhanced by almost an order of magnitude approaching 400 fT/Hz^{1/2} at the electromechanical resonance, as shown in Figure 6b [43]. Based on the giant resonance ME coupling coefficient in 1-1 type ME laminate, a superhigh resonant magnetic field sensitivity close to be 135 fT (see Figure 6c) was further obtained by Chu et al. [8], which indicates great potential for 1-1 type ME composites in the field of eddy current sensing, space magnetic sensing and active magnetic localizing [8][61]. In 2018 Turutin et al. reported a new ME composite consisting of the y + 140° cut congruent lithium niobate piezoelectric plates with an antiparallel polarized “head-to-head” bidomain structure and magnetostrictive material Metglas [44]. Based on this 2-2 ME bimorph, the equivalent magnetic noise spectral density was only 92 fT/Hz^{1/2} and the directly measured resolution was found to be 200 fT at a bending resonance frequency of 6862 Hz (see Figure 6d), but one should note that the bandwidth of resonant ME sensors is normally below 1 kHz due to the high mechanical quality factor, which is a major limitation facing practical engineering applications [8][44][62]. It should however be noted that resonant ME sensors are greatly limited by the narrow bandwidth and specifically suited applications need to be considered.

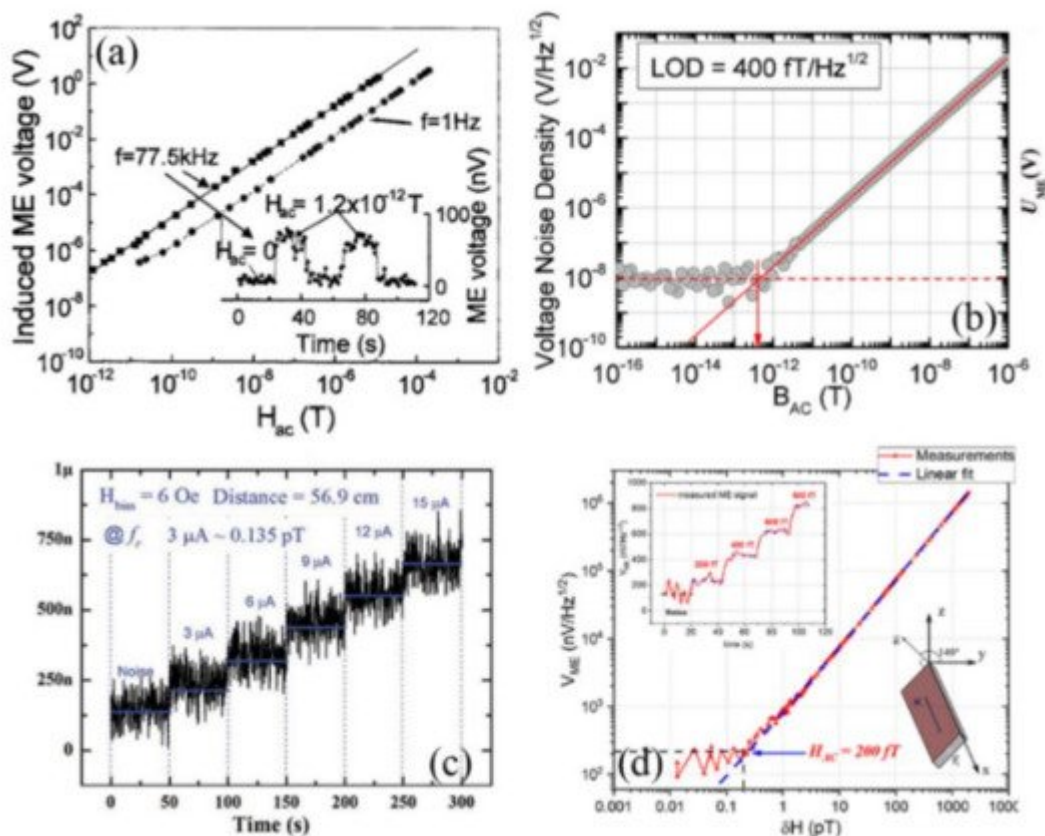


Figure 6. (a) Magnetic field detection limit measurements at frequencies of $f = 1 \text{ Hz}$ and $f = 77.5 \text{ kHz}$ (resonance condition), respectively [29]; (b) The measurement of LOD for MEMS ME sensor [43], (c) for 1-1 typed ME sensor [8] and (d) for a 2-2 ME bimorph [44].

3.3. DC Magnetic Sensor

DC or quasi-static magnetic sensors are promising for magnetic anomaly detection uses, such as geomagnetic navigation, metal detection and magnetic medical diagnosis, etc. Early in 2011, Gao et al. demonstrated the excellent detection ability for DC field using 2-1 ME composite [31]. As shown in Figure 7a,b, the magnetic resolution was found to be 4 nT and 1 nT when driving the composite at non-resonant frequency and resonance frequency, respectively [31]. In 2013, Nan et al. reported a self-biased 215 MHz magnetoelectric NEMS resonator consisting of an AlN/(FeGaB/Al₂O₃) multilayered heterostructure (Figure 7c), for ultra-sensitive DC magnetic field detection [51]. An ultra-sensitive detection level starting from 300 picoTesla was obtained experimentally (Figure 7d) [51]. The RF NEMS magnetoelectric sensor is compact, power efficient and readily integrated with CMOS technology, however, the measurement of the resonance frequency and the admittance spectrum is not technologically convenient. Li et al. then further proposed to monitor the reflected output voltage from the ME resonator directly [26]. The optimized detection sensitivity was determined as 2.8 Hz/nT for AlN/FeGaB resonator. An ultra-high frequency (UHF) lock-in amplifier and a directional coupler were used to apply and test the RF signal of this resonator. And the final limit of detection was measured to be around 0.8 nT.

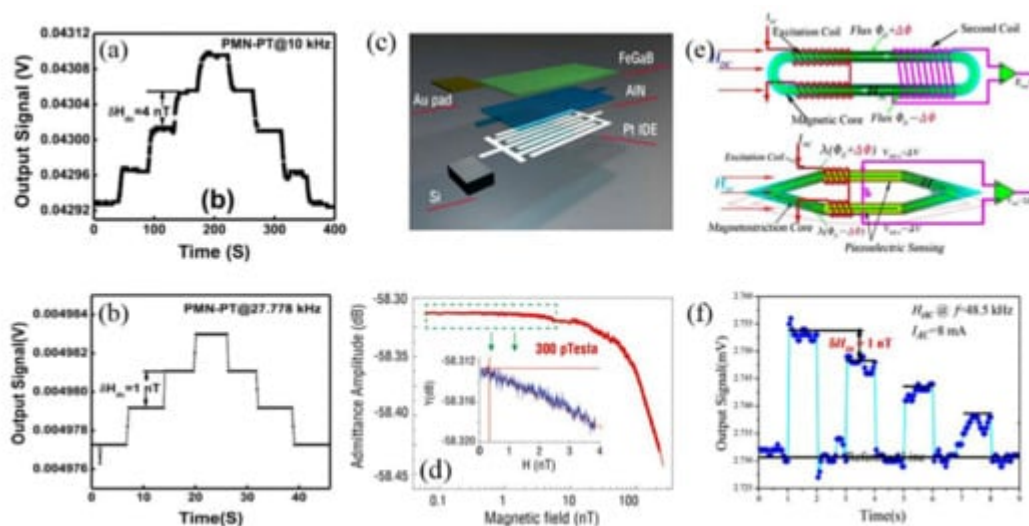


Figure 7. The measurement of LoD for Metglas/PMN–PT ME laminate at (a) $f = 10$ kHz and (b) resonance frequency of 27.778 kHz [31]. (c) Schematic representation and (d) the measurement of LoD for NEMS AlN/(FeGaB/Al₂O₃) multilayered heterostructure [51]; (e) Schematic representation of the conventional flux gate sensor and the proposed ME flux gate sensor [63]; (f) The measured results for DC magnetic field resolution [63].

Using the nonlinear resonance magnetoelectric effect in ME composites, Burdin et al. fabricated a planar langatate-Metglas structure and employed the third harmonics of the output signal to measure the DC magnetic field as low as 10 nT [64]. In addition, a broad dynamic range from ~ 10 nT to about 0.4 mT was also successfully obtained using the nonlinear ME effect [65]. More recently, Chu et al. proposed a shuttle-shaped, non-biased magnetoelectric flux gate sensor (MEFGS) for DC magnetic field sensing enlightened by the design of conventional flux gate sensor [63]. Figure 7e shows both the schematic of typical flux gate sensor and the proposed magnetoelectric flux gate sensor. The flux gate sensor based on Faraday's Law of Induction is composed of a racetrack type magnetic core surrounded by an excitation (first) coil and a detection (second) coil. With respect to MEFGS, a similar differential structure, which can produce a longitudinal-bending vibration when applying a DC

field, can reject in-phase vibration noise and enhance the out-of-phase ME voltage signal simultaneously [54]. We note here that in [54] the authors found that a ME flux gate sensor excited under a non-resonant high frequency field could perform better detection ability. As shown in Figure 7f, the relative change of the ME voltage output signal in response to a LOD of 1 nT is around 0.2% and the output signal can return to the reference level during the repeated test cycles when choosing a non-resonant frequency of 48.5 kHz [63].

Performance summary of some typical magnetoelectric sensors was given in Table 2. Table 3 further compares the LoD of passive ME sensors with some commercially available magnetometers, i.e., magnetoresistive sensors, giant magneto-impedance sensors, fluxgate sensors, optically pumped magnetometers and SQUID magnetometers. As it can be seen in Table 3, ME sensor shows comparable and competitive performance with these products. Specifically, the low power consumption and high detection ability are significant advantages for ME sensors, while vibration interference still now greatly limits the engineering applications. On the other hand, piezoelectric materials are normally susceptible to the working temperature and the temperature stability of ME sensors is also a critical issue. For example, Burdin et al. compared the temperature dependence of the resonant magnetoelectric effect in several kinds of ME composites and showed that the widely studied PZT-Metglas ME sensor can only work in a narrow temperature range of 0 °C to +50 °C [66].

Table 2. Performance summary of typical magnetoelectric sensors.

	Composition	Working Mode	Sensing Mode	LoD
Low-frequency magnetic field sensing	Metglas/Mn-PMNT [67]	Longitudinal vibration (Multi-L-T)	Passive sensing	0.87 pT/√Hz @ 30 Hz
	Metglas/PMN-PT [33]	Longitudinal vibration (Multi-push-pull)	Passive sensing	5.1 pT/√Hz @ 1 Hz
	Metglas/PMN-PZT [55]	Longitudinal vibration (L-T)	Active Modulation	33 pT/√Hz @ 0.1 Hz
Resonant magnetic field sensing	Metglas/ LiNbO3 [44]	bending mode	Direct Sensing	92 fT/√Hz
	FeCoSiB/(Pt)/AlN [43]	bending mode	Direct Sensing	400 fT/√Hz

	Metglas/PMN-PZT [8]	Longitudinal vibration (L-T)	Direct Sensing	123 fT/√Hz
	langatate-Metglas [64]	bending mode	Nonlinear ME effect	10 nT
DC magnetic field sensing	Metglas/PMN-PZT [9]	Longitudinal vibration (L-T)	Linear ME effect	1 nT
	FeCoSiB/(Pt)/AlN [26]	Lateral vibration	Delta-E effect	0.8 nT
	FeCoSiB/(Pt)/AlN [51]	Lateral vibration	Delta-E effect	0.4 nT

Table 3. Performance Comparison with commercially available magnetometer for 1 Hz magnetic field sensing.

Magnetometer	Working Temperature	Power Consumption (mW)	Typical Size	LoD@1Hz (pT/√Hz)	Limitations
ME sensor [33]	0 °C to +50 °C ①	<1	80 mm × 10 mm @ ME composites	5.1	Vibration interference
Magnetoresistive sensor ②	−40 °C to +125 °C	~0.02	6 mm × 5 mm × 1.5 mm @ sensing element	100	Low sensitivity
Giant magneto-impedance sensor ③	−20 °C to +60 °C	75	35 mm × 11 mm × 4.6 mm	15–25	Low sensitivity

@ sensing element					
Fluxgate magnetometer ④	−40 °C to +70 °C	350	∅100 mm × 125 mm @ system size	2–6	Power consumption
Optically pumped magnetometer ⑤	−35 °C to +50 °C	>12,000	175 cm × 28 cm × 28 cm @ system size	4	Complex setup
SQUID magnetometer [68]	<−196 °C	>1000	12.5 mm × 12.5 mm @ chip size	<0.005	Cooling

① Estimated from the data in ref. [64]; ② Based on commercial product TMR9001 in MultiDimension Technology Co., Ltd. (Zhangjiagang Free Trade Zone, Jiangsu Province, China); ③ Based on commercial product MI-CB-1DH in AICHI STEEL CORPORATION (Tōkai city, Aichi Prefecture, Japan); ④ Based on commercial product Mag03 from Bartington Instruments Ltd (Witney, Oxon, OX28 4GG United Kingdom).; ⑤ Based on commercial product G882 marine magnetometer from GEOMETRICS, INC (San Jose, CA, USA).

4. Engineering Applications of ME Sensors

As we summarized in [Table 2](#) and [Table 3](#), ME sensors show competitive performance with commercial optically pumped magnetometers, giant magneto-impedance sensors and fluxgate magnetometers. In this regard, a large number of works that utilize ME sensors for magnetic field sensing have been published and various applications have been implemented.