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Giant low-frequency magnetoelectric torque (MET) effect in polyvinylidene-fluoride (PVDF)-based MET device*

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A polyvinylidene-fluoride (PVDF)-based magnetoelectric torque (MET) device is designed with elastic layer sandwiched by PVDF layers, and low-frequency MET effect is carefully studied. It is found that elastic modulus and thickness of the elastic layer have great influences on magnetoelectric (ME) voltage coefficient (α_{ME}) and working range of frequency in PVDF-based MET device. The decrease of the modulus and thickness can help increase the α_{ME}. However, it can also reduce the working range in the low frequency. By optimizing the parameters, the giant α_{ME} of 320 V/cm·Oe (1 Oe = 79.5775 A·m^{-1}) at low frequency (1 Hz) can be obtained. The present results may help design PVDF-based MET low-frequency magnetic sensor with improved magnetic sensitivity in a relative large frequency range.

Keywords: magnetoelectric torque effect, piezoelectric, ME voltage coefficient

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1. Introduction

The magnetoelectric (ME) effect is the phenomenon of inducing magnetization/electric polarization by applying an electric/magnetic field.[1] The materials or structures showing ME effect have wide applications in magnetic field detection, information storage, energy harvesting, etc.[2–6] The strength of the ME effect can be evaluated by the ME voltage coefficient α_{ME} ($\alpha_{\mathrm{ME}} = \delta E/\delta H$, where H is the applied magnetic field, and E is the electric field). The larger α_{ME} means the higher magnetic field sensitivity, which motivates the search for ME materials or structures with larger α_{ME}.

ME effect was first discovered in single phase $\mathrm{Cr_2O_3}$,[7] however, the α_{ME} was very weak ($\sim$ 10 mV/cm·Oe) and appeared at extremely low temperature.[2] Then in 1972, particle composites consisting of magnetostrictive and piezoelectric materials were presented at Philips lab. But after decades of development, preparation technology needs to improve to make the preparation easy enough.[8] Until 2001, Ryu *et al.*[9,10] successfully fabricated magnetostrictive-piezoelectric laminate composites. At the same time, the α_{ME} could reach as high as 4680 mV/cm·Oe at room temperature, which is about 36 times higher than the best value reported. In recent years, vast investigations have been conducted on the laminated ME composites for their excellent performances with larger values of α_{ME}.[11–17] The values of α_{ME} for laminated ME composites could be as high as $\sim$ 20 V/cm·Oe at the low frequency and 1800 V/cm·Oe at the resonant frequency, respectively.[18] Besides the laminated ME composites, new ME structures with the new ME coupling mechanism were designed.[19,20] In 2008, a new type of ME device composed of piezoelectric materials and magnets was proposed by Xing *et al.*[20] The ME coupling originates from the interaction between magnetic torque and piezoelectricity, which is named magnetoelectric torque (MET) effect. The MET devices can reach tens to hundreds V/cm·Oe at the low frequency and as high as several thousand V/cm·Oe at the resonant frequency.[21,22] Compared with ME devices, the MET devices have large values of α_{ME} at the low frequency, which exhibits the potential in the sensitive detection of low frequency magnetic field. Xing and Xu have investigated the low frequency MET effects in $\mathrm{Pb(Zr_{1-x}Ti_x)O_3}$ (PZT)-based MET devices and obtained the ME voltage coefficient equation: $\alpha_{\mathrm{ME}} = -3Md_{31}/4Wh^2\varepsilon_{33}^{\mathrm{T}}B$.[21] From this equation, one can clearly see that α_{ME} can be tuned by the $d_{31}/\varepsilon_{33}^{\mathrm{T}}$. For a PZT-based MET device, the α_{ME} of low frequency is about 100 V/cm·Oe and 2100 V/cm·Oe at the resonant fre-

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quency, which is comparable to the scenario of laminated ME composites.[22] In order to increase α_{ME} at low frequency, the value of $d_{31}/\varepsilon_{33}^{\mathrm{T}}$ needs increasing. As is well known, the $d_{31}/\varepsilon_{33}^{\mathrm{T}}$ of polyvinylidene fluoride (PVDF) is about 1.79, which is nearly 20 times larger than that of PZT. Therefore, a giant α_{ME} can be expected in a PVDF-based MET device in theory. However, PVDF is a soft material with a Young's modulus of about 2 GPa, which cannot transfer the strain easily.[23,24] Therefore, an elastic layer with large Young's modulus is needed to support PVDF for effectively transferring the strain. In order to achieve the optimized output (i.e., α_{ME} and the working range of frequency) of the MET devices, the influences of Young's modulus and thickness of the elastic layer should be carefully studied.

In this paper, we design an MET device with elastic layer sandwiched by PVDF layers. The low-frequency MET effect is carefully investigated. Reducing the modulus and thickness can help increase the value of α_{ME}. However, it also reduces the working range in the low frequency. By optimizing the parameters, including modulus and thickness of elastic layer, number of magnets, the giant α_{ME} of 320 V/cm·Oe at low frequency can be obtained.

2. Experiment

The MET device composed of piezoelectric layers, elastic layer and magnets (NdFeB) is shown in Fig. 1(a). The elastic layer is metallic, while the layers on and beneath the elastic layer are both piezoelectric. One end of the device is clamped, and the other end is free with magnets loaded on it, which is a cantilever beam structure. The symmetrical structure of the two piezoelectric layers can avoid the noise induced by the fluctuation of the environmental temperature.[25] The magnetization directions of the magnets are both along the direction of the layer thickness, and the direction of the applied alternating current magnetic field H_{ac} is perpendicular to the magnetization directions of the magnets. Therefore, the applied magnetic field H_{ac} interacts with the magnetic moment m of the magnet inducing a magnetic torque τ. This torque will bend the layers. Due to the direct piezoelectric effect, the piezoelectric layers will induce an ME voltage output.

Our samples were composed of PVDF layers (32 mm×6 mm×0.1 mm in size), metal layer (32 mm× 6mm in size) and magnets. To investigate how the elastic layer's thickness affects the MET effect, we have prepared eleven elastic layers with different metal thickness values: 0 mm, 0.01 mm, 0.02 mm, 0.03 mm, 0.04 mm, 0.05 mm, 0.06 mm, 0.08 mm, 0.1 mm, 0.15 mm, and 0.2 mm. To observe the effects of the metal elastic modulus on the MET effect, we have chosen three kinds of metals as the elastic layer, which are stainless steel, copper and aluminum, and their moduli of elasticity are 200 GPa, 100 GPa, and 70 GPa, respectively. The dimensions of the two permanent magnets NdFeB are both Φ8 mm×2 mm, and also the residual flux density of the magnet $B_{\mathrm{r}} = 1.2$ T.

For the MET samples, two PVDFs were polarized in the opposite thickness directions. PVDF and metal layer were bonded together by glue with conducting silver paste. In the experiment on each thickness value of the metal, we used at least three samples to ensure the accuracy and repeatability.

The measuring system was illustrated in Fig. 1(b). The MET device was located in the Helmholtz coil which was used to supply an AC magnetic field $H_{\mathrm{ac}} = 0.5$ Oe. We measured the ME voltage output by an SR7270 DSP lock-in amplifier.

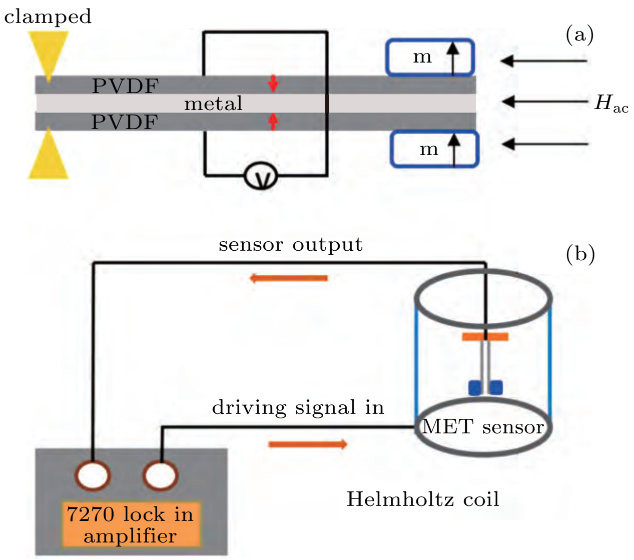

Fig. 1. (color online) (a) Sandwiched structure of the device and (b) measuring system for the MET device.

3. Results and discussion

Figure 2 shows a typical frequency dependence of α_{ME} for one MET device which is composed of PVDF, steel, and magnet. The sample parameters are listed in the figure. Each of the two PVDF layers has a size of 32 mm×6 mm×0.1 mm and the steel size is 32 mm×6 mm×0.01 mm. Each of two magnets is Φ8 mm×2 mm in size. It is clearly seen from the figure that α_{ME} shows a resonant peak with increasing frequency. The resonant frequency f_{r} is about 9.3 Hz. The α_{ME} keeps stable when the frequency is below the one where $\Delta\alpha_{\mathrm{ME}}/\Delta f \cdot \alpha_{\mathrm{ME}}$ is below 10%, which is defined as the low frequency region (quasi-static region ΔF). The α_{ME} can reach 4439 V/cm·Oe at the resonance frequency. It is noted that the MET device has a giant α_{ME} of 305 V/cm·Oe at the low frequency, which is much higher than the one that the laminated composites-based device has.

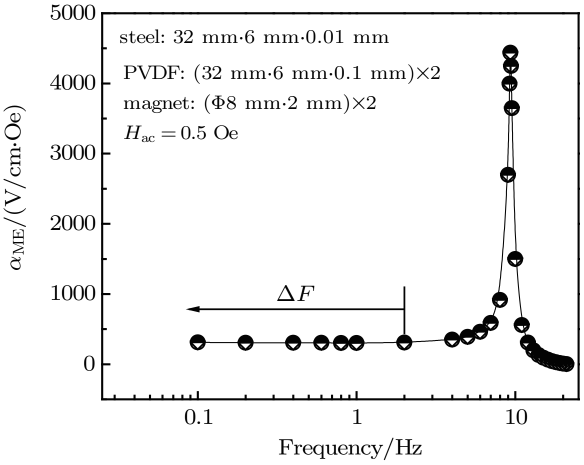

Fig. 2. Typical frequency dependence of α_{ME}.

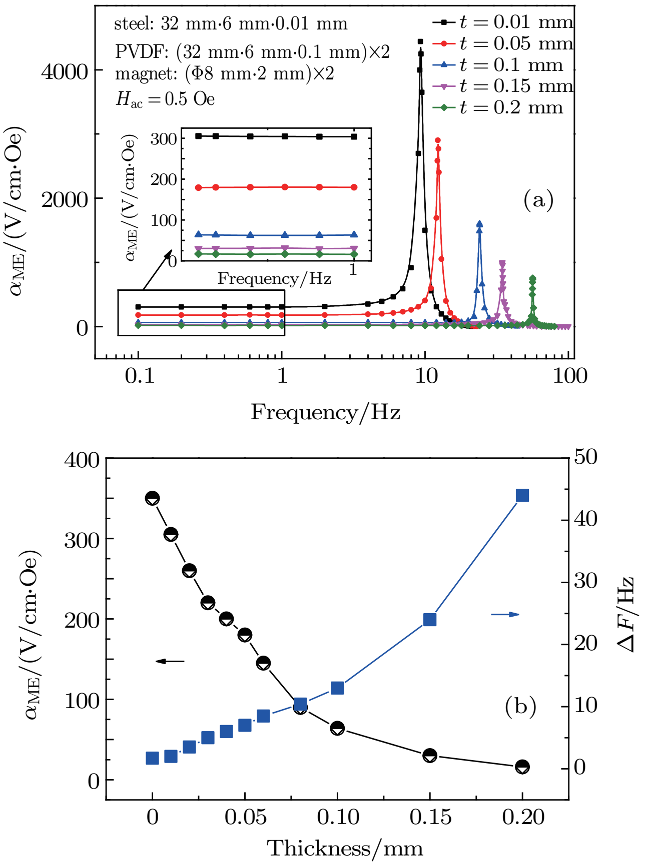

Fig. 3. (color online) (a) Frequency dependence of α_{ME} for different steel thickness values. The inset shows the variations of α_{ME} with frequency in the quasi-static region for different steel thickness values. (b) Steel thickness dependence of α_{ME} and ΔF in the quasi-static region.

To investigate the influence of the thickness on MET effect, the values of α_{ME} are measured for MET devices with metal layers of five different thickness values. Figure 3(a) shows the parameters of the MET devices and the frequency dependence of α_{ME} for different thickness values of metal layer. It can be seen that the resonant peaks of α_{ME} shift to the higher frequency region with a value ranging from 9.3 Hz to 56.3 Hz. Moreover, the value of α_{ME} in the quasi-static region decreases with the increase of metal thickness as shown in the inset of Fig. 3(a). It declines from 305 V/cm˙Oe to 17 V/cm·Oe. In Fig. 3(b), we choose eleven kinds of steel thickness values to make samples and we record the experimental values at the quasi-static frequency (∼ 1 Hz). One black curve with circular data points shows the relationship between the steel thickness and the α_{ME}. We can see that as the thickness of steel increases, the value of α_{ME} decreases from 350 V/cm·Oe to 16 V/cm·Oe. To achieve a large α_{ME}, the thickness of the metal is better to select a small one. However, the other blue curve with rectangular data points in this graph shows that when the thickness of the steel decreases, the scope of the low stable quasi-static frequency of the device decreases with ΔF changing from the 44 Hz to 1.7 Hz.

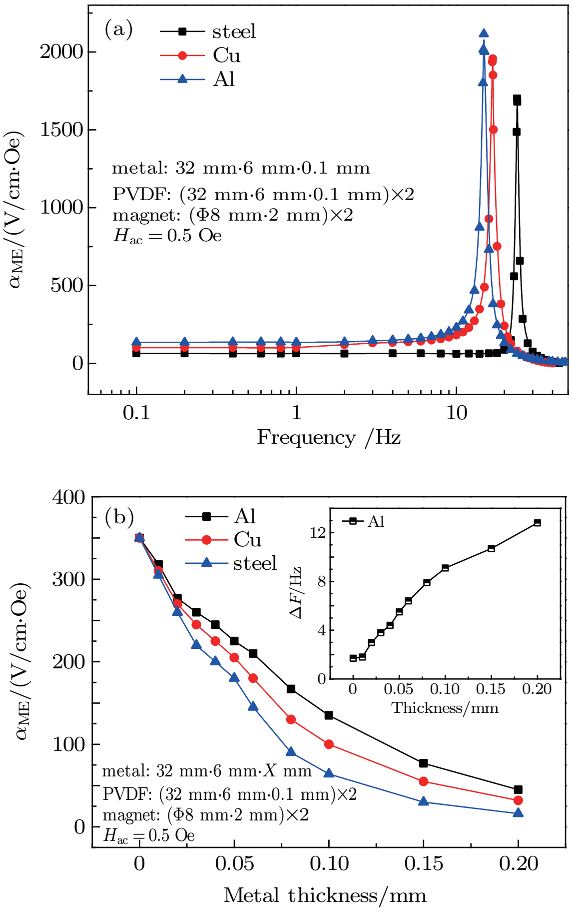

Fig. 4. (color online) (a) Frequency dependence of α_{ME} for different metals. (b) Metal thickness dependence of α_{ME} in quasi-static region for different metals. The inset shows thickness dependence of ΔF in quasi-static region for aluminum.

To observe how the elastic modulus of the metal will affect the MET effect, we choose three kinds of metals as the elastic layer. They are steel, copper and aluminum. The MET samples we prepared have the same parameters which are given in Fig. 4(a). In this figure, we can see the curves showing the frequency dependence of α_{ME} for different metals. With the increase of the elastic modulus of metal, the resonant peak of α_{ME} shifts from 14.9 Hz to 29 Hz while the values of α_{ME} in the quasi-static region for different metals decrease from 135 to 63 V/cm·Oe. Figure 4(b) shows that as the metal thickness increases, the values of α_{ME} in quasi-static region for different metals decrease. In particular, for the same thickness value of different metals, we can know that the values of α_{ME} decrease with elastic modulus of metal increasing. With the same thickness, the MET device has an aluminum layer showing the highest values of α_{ME} for three metals. So the thickness dependence of ΔF for aluminum is necessary for the importance of the working frequency range. In the inset of Fig. 4(b), the ΔF in quasi-static region increases with the increase of thickness for aluminum, and it increases from about 1.7 Hz to 12.8 Hz with the thickness of aluminum changing from 0 mm to 0.2 mm, but is lower than that for steel. It is known that the elastic modulus of metal is much larger than that of PVDF. When the thickness of the metal or the elastic modulus of metal increases, the torque bending the beam becomes much more difficult, which reduces the α_{ME}.

When the material and the size are selected, it is possible to adjust the magnets to obtain the larger α_{ME}. Some experiments are carried out to testify this. We choose the steel as the elastic layer, and the thickness of the steel is 0.1 mm. Then we increase the number of magnets by two from two to ten. The parameters of the MET device are shown in Fig. 5. From this figure, we can conclude that the experimental result presents a linear relationship which exists between the magnet's residual magnetism and the α_{ME}. According to this, the α_{ME} can reach infinity in theory.

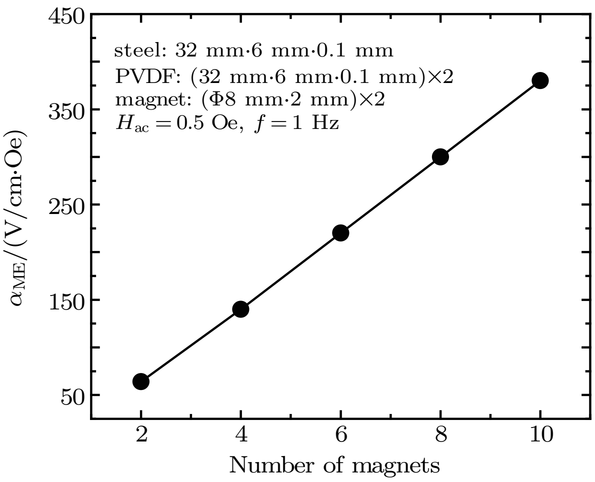

Fig. 5. Magnet number dependence of α_{ME}.

According to the literature, the product of Young's modulus, tensile strain and thickness determines the balance force.[26] With a constant magnetic field applied, the bending force induced by magnets is fixed. For a metal layer with the known Young's modulus, if its thickness increases, then a small tensile strain is needed to balance the bending force, thereby leading to a slight tensile strain of PVDF layers. On the other hand, when the metal layer has a certain thickness, the increase of the Young's modulus also means that a small tensile strain is needed to balance a certain bending force. It also results in a small tensile strain in PVDF layers. In these two conditions, PVDF layers generate a small quantity of charge according to the piezoelectric effect. It is universally received that the small quantity of charge directly reduces the intensity of polarization, producing a weak electric field. From the definition of the α_{ME}, the weak electric field obviously reduces the α_{ME}, with a constant magnetic field applied. Therefore, the increase of Young's modulus or thickness of the metal layer induces α_{ME} to decrease. However, we can tune the Young's modulus and the thickness of the elastic layer to produce a large tensile strain to balance the bending force which can make a giant α_{ME}. For example, when the aluminum is chosen as the elastic layer and its thickness is chosen to be 0.01 mm, the α_{ME} of low frequency can reach about 320 V/cm·Oe. In addition, we can adjust the magnets to increase the α_{ME}. Therefore, we can take the overall conditions into consideration to design the MET device for obtaining a required value of α_{ME}.

4. Conclusions

An MET device with elastic layer sandwiched by PVDF layers is designed, and the MET effect at low frequency is carefully studied. It is found that the elastic modulus and thickness of elastic layer have great influences on the α_{ME} in PVDF-based MET device. The value of α_{ME} decreases with the increase of thickness and elastic modulus of metal, but at the same time, the low stable quasi-static frequency range increases. So we should select a suitable thickness of the elastic layer to obtain a giant α_{ME} of the MET device according to the working range of frequency. In addition, the enhancement of magnets for the device also contributes to getting a large α_{ME}. The results offer a candidate for a low frequency magnetic sensor based on MET device.

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