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Giant electric-field-induced magnetization in a magnetoelectric composite at high frequency

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Electric-field-induced magnetization (EIM) characteristics are investigated in a small $0.68[Pb(Mg_{1/3}Nb_{2/3})O_3]0.32PbTiO_3$/Terfenol-D bilayered composite by using double coils in a wide frequency range of 10–700 kHz. The EIM behaviors are strongly dependent on driving electric field frequency, bias magnetic field, and angle θ between the bias magnetic field and polarization direction. The magnetoelectric coefficient at electromechanical resonance frequency of 185 kHz reaches up to 3.1×10^{-7} s/m, which is $\sim$60 times higher than the values at nonresonance frequency. The EIM variations with the bias magnetic field exhibit hysteresis, forming EIM hysteresis loops caused by the magnetostrictive hysteresis from Terfenol-D. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2998699]

Multiferroic materials have drawn much research interest because of their simultaneous ferroelectric/antiferroelectric and ferromagnetic/antiferromagnetic orders.[1,2] Magnetoelectric (ME) coupling may arise directly between the two order parameters or indirectly via strain. Single-phase multiferroic materials have difficulties in device application due to their low Curie temperature and weak coupling between electric polarization and magnetization[2] except $BiFeO_3$ (Ref. 3). Alternatively, composites enjoy a stronger effective strain-mediated ME coupling owing to their giant product property of the piezoelectric and magnetostrictive effects at room temperature.[4] Among them, giant ME effect has been found in the composites containing $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3-xPbTiO_3$ (PMN-PT) with high piezoelectric constant[5,6] and Terfenol-D[5,7–10] with high magnetostrictive coefficient, especially in the laminate composites.

The ME coupling is characterized by magnetic-field-induced electric (MIE) polarization or electric-field-induced magnetization (EIM). Relatively, the ME effect can be expressed by an equal strength $\alpha=(\partial P/\partial H)_E=\mu_0(\partial M/\partial E)_H$ (Ref. 11) and with two converse investigation methods. Up to date, most studies on the ME effect have focused on the MIE. Seldom report has been made on the magnetic response to an applied electric field. In fact, EIM has both physical interest[12] and technological applications such as coil-free electromagnet[5,13] and ME memory.[14] On the other hand, the MIE measurement commonly requires Helmholtz coils to generate a dynamic magnetic field.[8–10] The impedance of the Helmholtz coils is $Z_L=\omega L_s$, where L_s is the inductance of the coils, resulting in a difficulty to obtain high-frequency magnetic field. Thus, researchers commonly select large samples for their low electromechanical resonance frequency.[5,7–10] This is unfavorable to miniaturize devices. Whereas, dynamic electric field with high frequency is easily applied on the sample for $Z_c=1/\omega C_p$ during EIM measurement, where C_p is the capacitance of the sample. So far, several EIM measurement methods have been developed: (1) the induced magnetization of the sample under an applied electric field was examined with a vibrating sample magnetometer,[11,15] (2) a search coil wrapped around the composite sample acquired the induced magnetization,[5] and (3) a search coil on the sample surface measured the induced magnetization.[10,13] The two former methods are inconvenient and the later cannot characterize the induced magnetization from the whole sample. In this letter, we investigate the EIM characteristics in a small PMN-PT/Terfenol-D bilayered composite by using double coils, which can conveniently detect the EIM behaviors representing the whole sample and compare the ME coupling from different samples.

Terfenol-D and $0.68[Pb(Mg_{1/3}Nb_{2/3})O_3]-0.32PbTiO_3$ (001) plates were commercially supplied and bonded together by epoxy bonder. The piezoelectric coefficient ($d_{33,p}$) of the PMN-PT chip is about 1600 pC/N and the Terfenol-D strain ($\lambda_{\parallel}$) is 834 ppm under the magnetic field of 1000 Oe. The PMN-PT component was polarized in the thickness direction. The sample was set 4.03×4.14 mm^2 in rectangle and 0.7 mm and 0.6 mm in thickness for PMN-PT and Terfenol-D, respectively. The EIM measurement was performed with an induction method. The laminate sample was placed in a dc bias magnetic field H_{bias} with search double coils as shown as Fig. 1. The planes of the double coils were perpendicular to the magnetic field. Applying a sine electric field δE_{ac} (about 83 V/cm) on the PMN-PT plate by a signal generator, the piezoelectric strain is induced in PMN-PT due to the converse piezoelectric effect. Then the strain is acoustically transferred to Terfenol-D; as a result, an induced mag-

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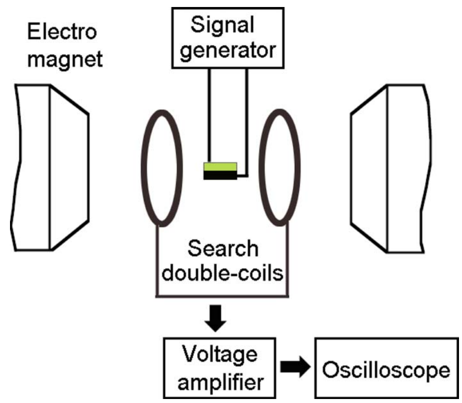

FIG. 1. (Color online) Schematic illustration of the testing setup for EIM measurement.

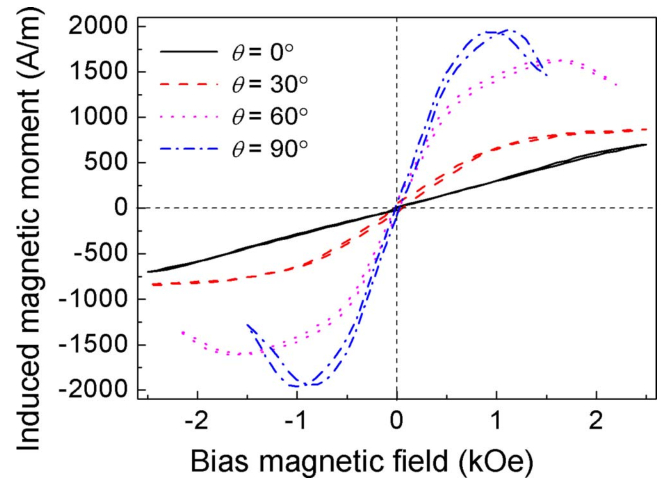

FIG. 3. (Color online) EIM hysteresis loops of the composite in different directions under electric field of 80 V/cm at 185 kHz.

netization δM_{ac} in Terfenol-D is produced due to the magnetoelastic coupling. The sine EIM in Terfenol-D induces a voltage in the double coils due to the Faraday effect. The coils are connected in series with relation $\phi=\sqrt{2}d$ for the highest magnetic flux, where ϕ is the diameter of the coils and d the distance between them. The double coils are connected to an amplifier, then to an oscillograph as shown in Fig. 1. The sample can rotate in the magnetic field. When the sample plane was perpendicular to the bias magnetic field (θ=0° or 180°), longitudinal behaviors were obtained, and when the sample plane was parallel to the bias magnetic field (θ=90° or 270°), transverse characteristics were measured. We also collected the phasic difference between the output magnetization and input sine electric field.

Figures 2(b) and 2(c) show the dependence of the transverse and longitudinal EIMs together with their phasic differences on frequency under an electric field of 83 V/cm at a bias magnetic field of 1000 Oe. The transverse EIM is stronger than the longitudinal EIM, which may be attributed to two reasons. The strain from the PMN-PT layer is in plane. When the measured EIM is in the same direction with the strain, the transverse EIM is obtained, where the magnetostrictive constant $d_{33,m}$ of Terfenol-D plays a key role in the magnetoelastic coupling. When the measured EIM is perpendicular to the strain direction, the longitudinal EIM is obtained, where $d_{31,m}$ plays a key role in the magnetoelastic coupling. Larger $d_{33,m}$ than $d_{31,m}$ (Ref. 16) leads to a stronger transverse EIM. Another reason is that there exists lower demagnetic field in the composite in the transverse direction. The transverse and longitudinal EIMs share similar characteristics. The EIM amplitude shows four strong resonant peaks, which correspond to the electromechanical resonance frequencies as shown in Fig. 2(a). Relatively, the phasic difference increases by $0.5\pi-\pi$ at each EIM resonance frequency, similar to the previous report.[10] EIM can be expressed as the product of piezoelectric and piezomagnetic effect, namely,

$$\alpha_M = \frac{\delta M}{\delta E} = \frac{\delta M}{\delta x}\frac{\delta x}{\delta E}, \tag{1}$$

where x is the strain. The phasic difference is between $0-\pi/2$ in each process of $\delta E/\delta x$ and $\delta x/\delta H$ analogous to the phasic difference between electric displacement D and electric field E in a dielectric material. Then the EIM phasic difference can reach π, agreeing with the experimental results of the phasic difference in $0.5\pi-\pi$ range around the EIM resonance frequencies.

Figure 3 shows EIM as a function of bias magnetic field under a driven electric field of 80 V/cm at 185 kHz. They show different behaviors at different measurement directions. In transverse direction (θ=90°), EIM increases up to a maximum at around H_{bias}=1000 Oe, then decreases with increasing H_{bias}, similar to that in the multilayer composite.[5] But when H_{bias} decreases, EIM does not reach the previous value, exhibiting a hysteresis phenomenon and forming three loops in the curve. This is caused by the magnetostrictive hysteresis from the Terfenol-D layer.[17] The maximum 1988 A/m obtained around 1000 Oe corresponds to the ME coupling

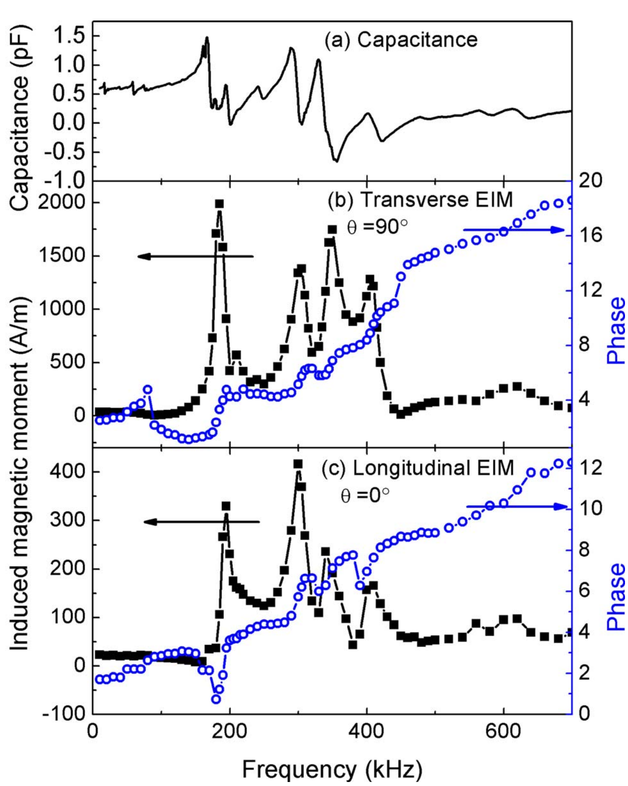

FIG. 2. (Color online) (a) Frequency dependence of composite capacitance. (b) Transverse and (b) longitudinal EIM responses and phasic differences of the laminate composite as functions of frequency under electric field of 83 V/cm and bias magnetic field of 1000 Oe.

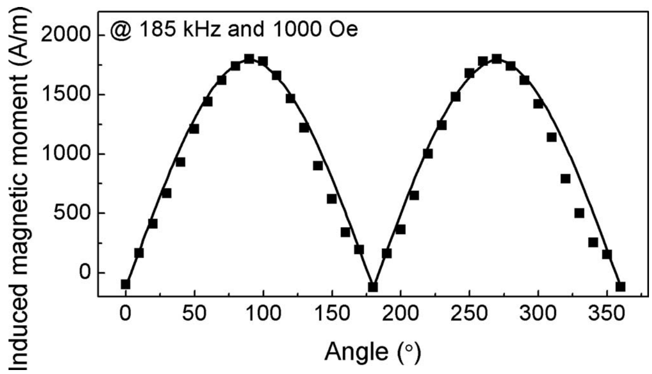

FIG. 4. Dependence of the EIM on the angle θ between the polarization and magnetic field. The line is fitted by using Eq. (3).

strength $\alpha=\mu_0(\partial M/\partial E)_H \sim 3.1\times 10^{-7}$ s/m, which is higher than that from the $La_{0.67}Sr_{0.33}MnO_3$ film on $BaTiO_3$ substrate[11] and the multilayered PMN-PT/Terfonel-D composite.[5] In longitudinal direction ($\theta=0°$), EIM cannot reach saturation even H_{bias} as high as 2.5 kOe because of the larger demagnetic field in the Terfenol-D layer. There exists weak magnetic hysteresis in this direction also. When $\theta=30°$ and $\theta=60°$, the curves are intervenient between those at 0° and 90°. There are not any responses to the bias magnetic field without electric field applied in the whole measurements. Therefore, the hysteresis phenomena are the results of ME coupling.

Then we investigate EIM anisotropy of the composite because of the different behaviors at different measurement directions. Figure 4 shows the EIM variation with angle θ measured by rotating the sample in the bias magnetic field of 1000 Oe under a driven electric field of 80 V/cm at 185 kHz. When the strain is transferred to Terfenol-D, the magnetostrictive constitutive equation in the Terfenol-D is[18]

$$\boldsymbol{S}=\boldsymbol{s}^H\boldsymbol{T}+\boldsymbol{d}_m\boldsymbol{H},$$

$$\boldsymbol{B}=\boldsymbol{d}_m^T T+\mu\boldsymbol{H}, \qquad (2)$$

where $\boldsymbol{B}$ and $\boldsymbol{H}$ denote the magnetic induction and magnetic field, respectively. $\boldsymbol{T}$ and $\boldsymbol{S}$ are the stress and strain, respectively. $\boldsymbol{s}^H$ is the elastic compliance coefficient at constant magnetic field, $\boldsymbol{d}_m$ is the piezomagnetic coefficient, and μ is the magnetic permeability at constant stress. The strain from the PMN-PT layer is in-plane; thus the transverse EIM is positive while the longitudinal EIM is negative relative to the piezomagnetic coefficients $d_{33,m}$ and $d_{31,m}$ (Ref. 16). Analogous to the MIE effect,[19] the EIM variation with angle θ can be written as

$$M(\theta) \sim A\sin^2\theta + B\cos^2\theta, \qquad (3)$$

where A and B are the parameters equal to the transverse EIM and longitudinal EIM, respectively. The experimental data with angle θ agree well with Eq. (3) as shown in Fig. 4.

In summary, EIM characteristics were studied in a small bilayered composite bonding a Terfenol-D alloy chip and a PMN-PT single-crystal chip with double coils. The highest ME coefficient 3.1×10^{-7} s/m has been obtained at electric field frequency of 185 kHz and bias magnetic field of 1000 Oe. The transverse EIM is higher than the corresponding longitudinal EIM due to the higher $d_{33,m}$ and lower demagnetic field in the transverse direction. The EIM hysteresis loops were observed, attributed to the magnetostrictive hysteresis from the Terfenol-D layer. The EIM at any angle θ has the intervenient behaviors between the transverse and longitudinal EIMs.

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