Converse magnetoelectric effects on heterotype electrostrain-piezopermeability composites

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By using the product effect of electrostrain and piezopermeability, a heterotype composite consisting of a bar of $Pb(Zr,Ti)O_3$ and a ring of MnZn ferrite with higher permeability has been developed, and its converse magnetoelectric effect has been investigated. Under a constant electric field of 5 kV/cm, about 20% electric-field-induced changes in permeability and impedance have been observed with the composite device over a wide frequency band. The electric-field dependence of the permeability and impedance shows a ferroelectric relaxation behavior. This composite is a candidate material for electrically controlled magnetic devices. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2979699]

Magnetoelectric (ME) and converse magnetoelectric (CME) effects are defined as magnetic field-induced change in polarization and electric-field-induced change in magnetization in materials, respectively.[1] Single-phase ME (or CME) materials, such as Cr_2O_3 or $Y(Ho)MnO_3$, are not recognized as suitable materials for practical use due to their low Curie temperatures and/or weak coupling between polarization and magnetization.[2–6] The laminated composites formed by piezoelectrics, such as $Pb(Zr,Ti)O_3$ (PZT) or $BaTi_{1-x}Fe_xO_3$, and magnetostrictive material, such as NiZn ferrite or $Tb_xDy_{1-x}Fe_{2-y}$, have been demonstrated to show superior ME performance owing to their giant product effect of the piezoelectric and magnetostrictive effects and negligible leakage currents, which can lead to loss of dielectric property.[7–9] However, there have been only a few reports on CME effects on layered composites.[10–12] However, the phenomenon is of fundamental and technological importance. It is desirable to synthesize a suitable CME material for use in electric field-controlled devices in order to alleviate the Joule heating and bandwidth limitation problems that are intrinsic in traditional electromagnetic transducers.

CME (ME) effects on laminated structure with alternating layers of ferroelectric and ferromagnetic phases are believed to originate from the variation in internal stress σ in the ferromagnet (ferroelectrics) in response to the electrostrain of the ferroelectrics (magnetostriction of the ferromagnet). For ferromagnets, especially those with higher permeability μ, it is verified that the permeability is much more sensitive to an applied magnetic field or a stress than its magnetostrictive coefficients.[13,14] In other words, the piezopermeability effect $(\partial\mu/\partial\sigma)$ is generally greater than their piezomagnetic effect $(\partial\lambda/\partial H)$,[7] thus more suitable for use as ferromagnet in fabricating CME composites, while magnets with higher magnetostriction are fit for ME devices.

On the other hand, graded ferroelectrics, which are capable of more efficient conversion of internal stresses into facial electric field compared to homogeneous ferroelectrics, are now extensively used to form efficient ME devices.[15–20] Thus, we have a conception of graded ferromagnets that are expected to form more superior CME composites. Some heterotypic ferromagnets can be considered as graded ones. In fact, a magnet with a closed magnetic circuit can generally show a larger piezopermeability effect than that with an open geometry.

In this letter, a heterotype composite consisting of a strip of PZT and a ring of manganese zinc (MnZn) ferrite with higher permeability is proposed. Moreover, observations of a large CME effect and the effect of electric-field-induced change in the impedance in the composite are presented.

In this study, MnZn ferrite with an initial permeability of 15 000 was employed as the piezopermeability material. The ferrite is in a shape of a ring 6 mm in height and 8 mm in diameter. A search coil of ten to 20 turns was wound and cemented with glue onto the ring. A strip of PZT with a length of 8 mm, width of 6 mm, and thickness of 2 mm was tightly fitted into the ferrite ring across the diameter as the electrostrain material, as shown in Fig. 1.

The coefficient of CME effects relating the electric field E and the permeability μ is given by

$$\alpha_\mu = \partial\mu/\partial E. \tag{1}$$

For a composite consisting of electrostrain and piezopermeability material, Eq. (1) can be expressed as

$$\alpha_\mu = \frac{\partial\mu}{\partial\sigma}\frac{\partial\sigma}{\partial E}, \tag{2}$$

where σ is the stress in the ferromagnet or the electrostrain phase, or between both, and $\partial\mu/\partial\sigma$ and $\partial\sigma/\partial E$ express the piezopermeability and the electrostrain effect, respectively. For a given ferroelectric with a stress perpendicular to the direction of its facial electric field, one should have $\partial\sigma/\partial E$

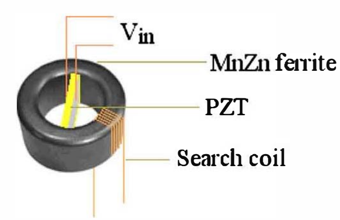

FIG. 1. (Color online) The schematics of the CME device composed of electrostrain material and heterotypic magnet with higher permeability.

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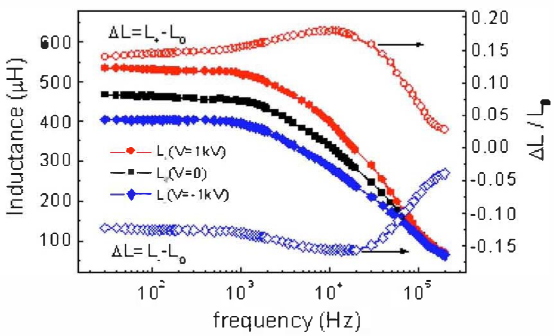

FIG. 2. (Color online) The inductance of the composite device for an electric field of 1 kV, 0 V, and −1 kV applied onto the PZT and the corresponding electroinductance as functions of excitation current frequency applied at room temperature.

$=\varepsilon_0\varepsilon_r/d_{31}$, where d_{31} is the piezoelectric coefficient and ε_0 and ε_r are the dielectric coefficients of vacuum and the piezoelectrics, respectively. Thus, one should have

$$\alpha_\mu = \frac{\varepsilon_0\varepsilon_r}{d_{31}}\frac{\partial\mu}{\partial\sigma}. \tag{3}$$

Equation (3) suggests that the CME effect for a composite with a given piezoelectric mainly relies on the piezopermeability of the magnet used.

Figure 2 shows the inductance of the heterotype device with and without a constant electric field applied to the PZT and the corresponding electroinductance as a function of the frequency of the current across the search coil. The ac current was supplied by an electrical bridge, which was used to measure the inductance and impedance of the device simultaneously. The electric-field-induced change in inductance, i.e., electroinductance, is defined as $\Delta L/L_0$, where L_0 is the inductance at zero electric field. Using the relation $L=N^2 v\mu_0\mu_{\mathrm{eff}}$, where N is the number of turns per unit length, v is the volume enclosed by the coil, and μ_{eff} is the effective permeability, one should have $\Delta L/L_0=\Delta\mu_{\mathrm{eff}}/\mu_{\mathrm{eff}\,0}$. That means electroinductance should be equal to electropermeability. As can be seen, under a constant positive or negative electric field that makes the PZT stretch and shrink, respectively, the composite shows a large positive or negative change in the inductance in a considerably wide bandwidth of the frequency from 30 to about 100 kHz. The peak value of electroinductance is located at about 15 kHz. In addition, the absolute value of the electroinductance effect under an electric field of −1000 V presents smaller than that under an electric field of 1000 V, possibly due to remanent polarization of PZT.

Besides the inductance, a coil always has a resistance and a distributed capacitance.[21] When an ac current is applied across the coil, those parameters are related to one another and to the signal frequency $f=\omega/2\pi$. Generally, the equivalent circuit of a coil suggests that the inductance and capacitance should be parallel and in serial with its resistance.[22] Thus, a change in inductance will cause a change in the impedance of the device by the relation given below

$$Z = \frac{\omega L R_p}{\sqrt{\omega^2 L^2 + R_p^2(1-\omega^2 C_p L)}}, \tag{4}$$

where R_p is the resistance of the device under a dc current and C_p is the capacitance in a static electric field. Figure 3 plots the impedance of the device as a function of frequency with the PZT under a positive, a zero, and a negative constant field, respectively, as well as the corresponding frequency dependence of electroimpedance, which is defined as $\Delta Z/Z_0$, where Z_0 is the impedance without a field applied on the PZT. It is found that the electroimpedance effect can also exist in a wide range of frequency from about 50 to over 1 MHz. The peak value of the electroimpedance is at about 5 kHz. Additionally, the electroimpedance effect is also influenced by the remanent polarization of the PZT.

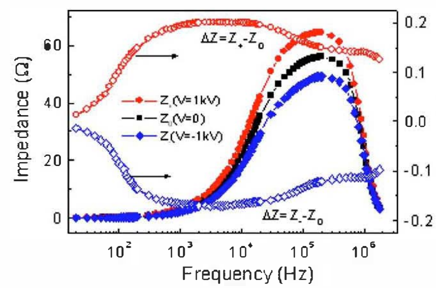

FIG. 3. (Color online) The impedance of the composite device with an electric field of 1 kV, 0 V, and −1 kV applied onto the PZT and the corresponding electroimpedance as functions of excitation current frequency applied at room temperature.

The dielectric relaxation of PZT is reflected in the data on inductance and impedance versus the field shown in Fig. 4. It should be noted that Fig. 4 actually exhibits the integrated result of the dielectric relaxation of the PZT and the magnetic hysteresis of the MnZn ferrite.

For a magnet under a stress, the initial permeability in a process of reversible magnetization can be expressed as

$$\mu_i = \frac{2\mu_0 M_s^2 l}{3\pi^2\delta\lambda_s\sigma} + 1, \tag{5}$$

where μ_0 is the permeability in vacuum, M_s is the saturated magnetization of the magnet, l and δ are the width and thickness of the wall of magnetic domains, respectively, λ_s is the saturated magnetostrictive coefficient, and σ is the internal

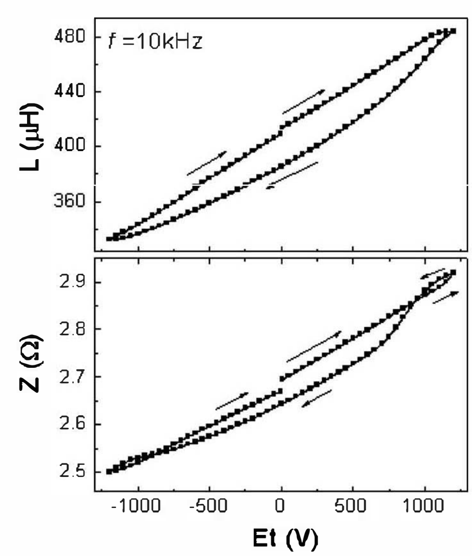

FIG. 4. The inductance (upper panel) and impedance (lower panel) of the composite at excitation current frequency of 10 kHz as functions of the electric field applied onto the PZT.

stress, which should include both internal and applied stresses. Differentiating Eq. (5) yields

$$\frac{\partial \mu_i}{\partial \sigma} = -\frac{2\mu_0 M_s^2 l}{3\pi^2 \delta\lambda_s \sigma^2}. \quad (6)$$

Additionally, considering the permeability dispersion induced by the magnetic relaxation for a magnet in an alternating field, one will have $\tilde{\mu}=\mu'-i\mu''$, where μ' and μ'' are elastic and viscous permeabilities, respectively. The effective permeability should be[23]

$$\mu_{\text{eff}} = \sqrt{\frac{\mu_i^2 + (\omega/\omega_c)^2}{1 + (\omega/\omega_c)^2}}, \quad (7)$$

where ω_c is the frequency of relaxation. Thus, in an ac field, $\partial\mu/\partial\sigma$ in Eq. (3) should be replaced by $\partial\mu_{\text{eff}}/\partial\sigma$. Collecting the results from Eqs. (3)–(8), one should have

$$\alpha_\mu = -\frac{\varepsilon_0 \varepsilon_r \mu_i^2}{d_{31}\sigma\sqrt{[1 + (\omega/\omega_c)^2][\mu_i^2 + (\omega/\omega_c)^2]}}. \quad (8)$$

For convenience, CME effect can also be simply expressed as the variation in the inductance of the search coil with changing the applied field, that is

$$\Delta L = N^2 v \mu_0 \alpha_\mu \Delta V/t, \quad (9)$$

where t is the thickness of the PZT and $\Delta V = t\Delta E$ is the voltage applied across the PZT. Since the piezoelectric coefficient d_{31} is generally negative in value, the inductance of the device should increase with increasing the voltage (Fig. 4) and decrease with increasing the frequency (Fig. 2). The present device is small in size, as well as cheap and easy to fabricate, so it is expected to be a smart and cheap room-temperature CME sensor that can be a complementarity of the one cent room-temperature ME sensor reported by Israel *et al.*[24]

In conclusion, magnets with high permeability and a closed magnetic circuit configuration show evidence for piezopermeability under an applied stress and are suitable for use in forming CME (or electromagnetic) devices. With a Mn–Zn ferrite of the characteristic above and a PZT composite, about 20% electric-field-induced changes in permeability (electropermeability) and impedance have been observed over a considerably wide frequency bandwidth under an applied electric field of 5 kV/cm at room temperature.

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