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Colossal magnetodielectric effect caused by magnetoelectric effect under low magnetic field

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Abstract. The colossal magnetodielectric effect is reported in Pb(Zr,Ti)O_3/Terfenol-D laminate composite under low magnetic field. When the composite is placed in an external a.c. magnetic field, magnetoelectric effect is produced, as a result, the dielectric properties of the Pb(Zr,Ti)O_3 is changed, i.e. magnetodielectric effect. Both the amplitude and resonance frequency change with the external magnetic field. The colossal magnetodielectric coefficient of 5 × 10^4% at low magnetic field of 20 Oe is achieved near the electromechanical resonance frequency.

Keywords. Composite materials; electrical properties; magnetodielectric effect; magnetoelectric effect.

1. Introduction

Recently, multiferroic materials have attracted much interest for their coexistence of ferroelectricity, ferromagnetism and ferroelasticity (Kimura *et al* 2003; Wang *et al* 2003; Hur *et al* 2004; Spaldin and Fiebig 2005; Eerenstein *et al* 2006; Weber *et al* 2006; Mamin *et al* 2007). The coupling between ferroelectric and ferromagnetic orders can produce some new effects, such as magnetoelectric (ME) and magnetodielectric (MD) effects. The MD effect is described as a change of dielectric constant in an applied magnetic field, i.e. MD $= \Delta\varepsilon(f)/\varepsilon(0, f)\times 100\% = \left[\varepsilon(H, f) - \varepsilon(0, f)\right]/\varepsilon(0, f)\times 100\%$, where $\varepsilon(H,f)$ and $\varepsilon(0,f)$ are the dielectric constants of the multiferroic materials at frequency f in and out of the external magnetic field, respectively. The MD effects have been studied mainly on single phases, which show dielectric change under an external magnetic field (Kimura *et al* 2003; Hur *et al* 2004; Lorenz *et al* 2004; Weber *et al* 2006; Mamin *et al* 2007). The possibility of tuning the dielectric constant by external magnetic field open perspectives for the basic understanding of the multiferroic materials and for the design of devices.

However, as the history of multiferroic research indicates, multiferroic materials in a single phase are known to be extremely rare (Nicola 2000; Prellier *et al* 2005). And the distinct change in the dielectric constant commonly occurs near the magnetic phase transition under external magnetic field (Kimura *et al* 2003; Prellier *et al* 2005), similar to the colossal magnetoresistance (Weber *et al* 2006). The dielectric anomalies near the phase transition resulted from strong competition and interplay among the charge, orbital and spin degrees of freedom (Mamin *et al* 2007). The MD effect is percentage value of several decades, and is much higher near the insulator–metal transition (Rairigh *et al* 2007) or incommensurate–commensurate transition (Kimura *et al* 2003). But the magnetic field needed for producing the MD effect is commonly as high as several teslas (Kimura *et al* 2003; Hur *et al* 2004; Lorenz *et al* 2004; Weber *et al* 2006; Mamin *et al* 2007; Rairigh *et al* 2007). Alternatively, composites have stimulated much scientific and technological interest because the multiferroic properties are easily obtained by combining the individual ferroic phases. The ME coupling in composites is much stronger resulting from the product property of both the phases even at room temperature in comparison with the single phase (Nan *et al* 2008). Analogously, the MD effect should be strong in the composites. On the other hand, the MD effect can be achieved through a combination of magnetoresistance and the Maxwell–Wagner effects without true ME coupling (Catalan 2006). Could the ME coupling result in a MD effect? Magnetoelastoelectric equivalent circuits are used to understand the ME coupling in the laminate composites (Dong *et al* 2003). It is easily concluded that magnetostrictive component under external magnetic field could change the dielectric properties of the piezoelectric component from the equivalent circuits. Therefore, the ME effect can result in a MD effect. In this paper, we choose a simple Pb(Zr,Ti)O_3/Terfenol-D laminate composite, in which a strong ME coupling is revealed (Zhou *et al* 2008), and study the colossal MD effect caused by the ME effect at much low a.c. magnetic field at room temperature.

2. Experimental

Terfenol-D and Pb(Zr,Ti)O_3 (PZT) were selected as magnetostrictive and piezoelectric components, respectively for

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their excellent magnetostrictive and piezoelectric properties. The PZT component was polarized along its thickness direction, d_{33}, of the PZT ceramic chip which is about 500 pC/N. The transverse Terfenol-D strain ($\lambda_{\parallel}$) is 860 ppm under the magnetic field of 700 Oe. The Terfenol-D and PZT tablets with the same diameter of 20 mm were bonded together by epoxy bonder. The thickness of the PZT and Terfenol-D was set to 1·5 mm and 3·5 mm, respectively.

The capacitance variation with frequency f_{E} of the sample was measured with an Agilent E4980A impedance analyser. A signal generator drives a power amplifier, then a solenoid to generate an a.c. magnetic field, $H = H_0 \cos 2\pi f_{\mathrm{M}} t$, where H_0 and f_{M} are the amplitude and frequency of the a.c. magnetic field, respectively. The sample was kept in the uniform magnetic field at the solenoid centre while performing the dielectric measurement to obtain the magnetocapacitance. When the sample plane was parallel to the magnetic field, the transverse magnetodielectric sensitivity was obtained; and when the sample plane was perpendicular to the magnetic field, the longitudinal magnetodielectric sensitivity was measured.

3. Results and discussion

We first studied the transverse magnetodielectric effect for the composite. Under applied external a.c. magnetic field $H = H_0 \cos 2\pi f_{\mathrm{M}} t$ with $f_{\mathrm{M}} = 10$ kHz and various amplitudes $H_0 = 0$, 1, 2, 3, 4, 6, 8, 12, 16, 20, 24 and 28 Oe, the dielectric constant of the composite was measured with an Agilent E4980A impedance analyser. The inset in figure 1(A) shows frequency f_E dependence of dielectric constant, in which three electromechanical resonance modes appeared around 38 kHz, 89·2 kHz and 113·1 kHz, and are assigned to the first-order bending resonance mode, the second-order bending resonance mode and radial resonance mode, respectively (Shi *et al* 2007; Zhou *et al* 2008). Figure 1(A) shows the enlarged patterns around the first-order bending resonance frequency to reveal the effect of the external a.c. magnetic field on the dielectric constant, in which the resonance frequency redshift occurs and the dielectric constant at peaks decreases first and then increases with H_0, exhibiting like a saddle. Thus, colossal MD effect is clearly present. The representative MD coefficient was summed in figures 1(B)–1(D), from which we know that the maximal MD can reach 104% when the magnetic amplitude H_0 is only 1 Oe. The maximal MD is as high as $5 \times 10^4\%$ at $H_0 = 20$ Oe. There exists similar characteristics around the other two resonance peaks. We defined relative changes of dielectric resonance frequency and dielectric resonance peak amplitude as

$$Rf_{\mathrm{E}} = \Delta f_{\mathrm{E}}/f_{\mathrm{E}}(0) \times 100\% = \left[f_{\mathrm{E}}(H) - f_{\mathrm{E}}(0)\right]/f_{\mathrm{E}}(0) \times 100\%,$$

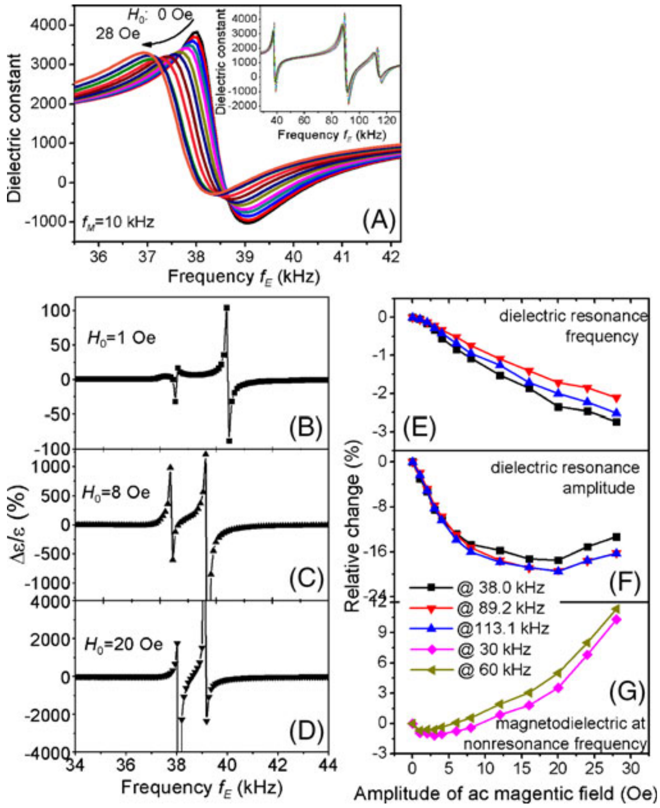

Figure 1. (A) Frequency f_E dependence of dielectric constants for the composite near the first-order bending resonance frequency under a.c. magnetic field with 10 kHz and various amplitudes parallel to the sample plane. The inset shows the corresponding dielectric constants in 28–130 kHz; (B)–(D) representative colossal magnetodielectric coefficients near the first-order bending resonance frequency under a.c. magnetic field with 10 kHz and amplitudes of 1, 8 and 20 Oe; (E) and (F) relative changes of resonance frequencies and resonance peak amplitudes with the a.c. magnetic field amplitude; (G) magnetodielectric coefficients at nonresonance frequencies of 30 and 60 kHz dependence on the a.c. magnetic field amplitude.

$$R\varepsilon' = \Delta\varepsilon'/\varepsilon'(0) \times 100\% = \left[\varepsilon'(H) - \varepsilon'(0)\right] \times 100\%,$$

where $f(H)$ and $f(0)$ are electromechanical resonance frequencies of the composite in and out of the external magnetic field; $\varepsilon'(H)$ and $\varepsilon'(0)$ are electromechanical resonance peak amplitudes of the sample in and out of the magnetic field, respectively. Then we summarize these changes for the three resonance modes in figures 1(E) and 1(F). The relative change of dielectric resonance frequency linearly decreases with increasing H_0, but the resonance amplitude represents U-shaped characteristics, i.e. decreases first and then increases. The relative change of the resonance amplitude reaches 19·5% at $H_0 = 20$ Oe even without any regard to

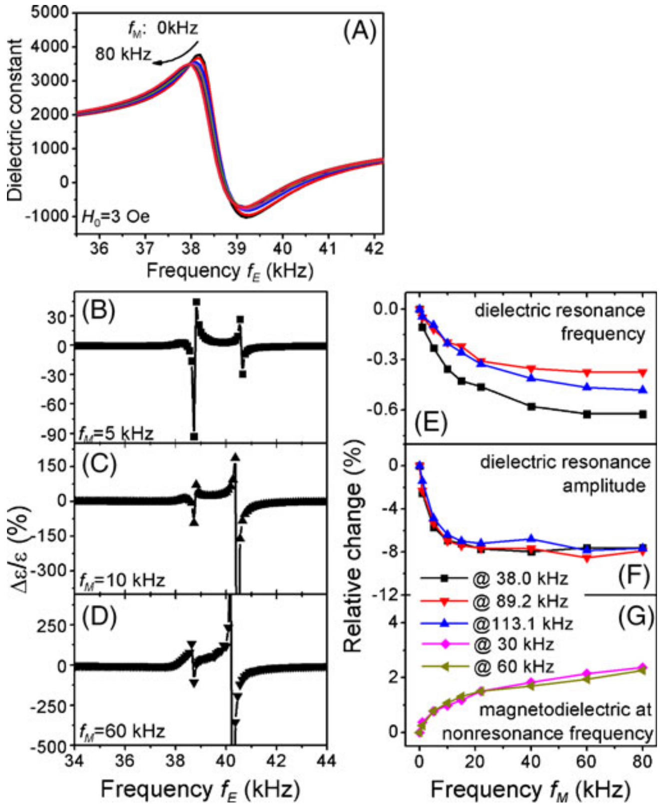

Figure 2. (A) Frequency $f_{\rm E}$ dependence of dielectric constants for the composite near the first-order bending resonance frequency under a.c. magnetic field with 3 Oe and various frequencies $f_{\rm M}$ parallel to the sample plane; (B)–(D) representative colossal magnetodielectric coefficients near the first-order bending resonance frequency under a.c. magnetic field with 3 Oe and frequencies of 5, 10 and 60 kHz; (E) and (F) relative change of resonance frequency $f_{\rm E}$ and resonance peak amplitudes with the a.c. magnetic field frequency $f_{\rm M}$; (G) magnetodielectric coefficients at $f_{\rm E} = 30$ and 60 kHz dependence on the a.c. magnetic field frequency $f_{\rm M}$.

frequency. MD coefficients at nonresonance frequency of 30 and 60 kHz are plotted in figure 1(G) for comparison. They decrease at first and then increase resulting from the characteristics in figure 1(A). The MD value reaches 10% at $H_0 = 28$ Oe. These values, even without considering the resonance frequency shift, are higher in comparison with the MD in single phase under much stronger magnetic field (Kimura *et al* 2003; Lorenz *et al* 2004).

Then we investigate the transverse MD effect under a.c. magnetic field with $H_0 = 3$ Oe and different frequencies $f_M = 1$, 5, 10, 15, 22, 40, 60 and 80 kHz. Figure 2(A) shows the frequency ($f_{\rm E}$) dependence of dielectric constant around the first-order bending resonance frequency. Colossal magnetodielectric effect is clearly present as shown in figures 2(B)–2(D) with the redshift of resonance frequency under a.c. magnetic field. The maximal MD reaches to 4×10^3% at $f_{\rm M} = 15$ kHz. Figures 2(E) and 2 (F) show the resonance frequency and amplitude changes with frequency

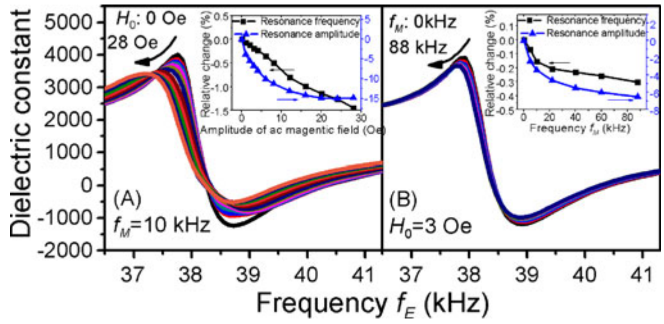

Figure 3. Frequency $f_{\rm E}$ dependence of dielectric constants for the composite near the first-order bending resonance frequency under external a.c. magnetic field: (A) with $f_{\rm M} = 10$ kHz and various amplitudes, and (B) with $H_0 = 3$ Oe and various frequencies $f_{\rm M}$ perpendicular to the sample plane. The inset in (A) shows relative change of resonance frequency $f_{\rm E}$ and resonance peak amplitude with the a.c. magnetic field amplitude. The inset in (B) shows relative change of resonance frequency $f_{\rm E}$ and resonance peak amplitude with the a.c. magnetic field frequency $f_{\rm M}$.

$f_{\rm M}$ of the a.c. magnetic field. The relative dielectric resonance frequencies and amplitudes decrease with increasing $f_{\rm M}$ first, and then become unchangeable. MD coefficients at non-resonance frequencies $f_{\rm M}$ of 30 kHz and 60 kHz increase with the magnetic field frequency and reach 2% at $f_{\rm M} = 80$ kHz as shown in figure 2(G).

The longitudinal MD effect shares similar characteristics with the transverse MD effect as shown in figures 3(A) and 3(B). The resonance peaks redshift occur with both the amplitude and frequency of the external a.c. magnetic field. But the relative changes are smaller than the corresponding values for the transverse MD effect.

The MD effect is relative with the ME effect. The ME polarization increases with the magnetic field (Shi *et al* 2007; Nan *et al* 2008), resulting in an enhanced MD coefficient as shown in figures 1(B)–1(D) and figures 2(B)–2(D). The longitudinal ME coupling is weaker than the transverse ME coupling (Zhou *et al* 2008), leading to a weak longitudinal MD coefficient. When applying magnetic field on the ME composite, the polarization was induced on the surface of sample due to the ME effect. The polarization impacts the dielectric response and change dielectric constant. The dielectric constant or the capacitance of the composites can be adjusted by the low applied magnetic field.

4. Conclusions

In summary, although MD effect can be achieved without true ME coupling, ME coupling can result in colossal MD effect in $Pb(Zr,Ti)O_3$/Terfenol-D laminate composite. The MD coefficients are sensitive to both the amplitude and frequency of the external a.c. magnetic field. The resonance frequency can be adjusted by an external a.c. magnetic field. The results are helpful for understanding the relationship between the MD and ME effects.

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