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Large magnetoelectric susceptibility: The fundamental property of piezoelectric and magnetostrictive laminated composites

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The magnetoelectric (ME) susceptibility (α_{me}) is the fundamental property that describes the coupling between the polarization and magnetization of a ME media. It is a complex quantity (α_{me}^*) which has rarely been studied. Here, we report investigations of the ME susceptibility for various ME laminated composites, which demonstrates that α_{me} is on the order of 10^{-7} s/m for these materials, which is dramatically larger than that of single phase materials. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2405015]

Magnetoelectricity is the interaction between the polarization and magnetization subsystems of a solid. Magnetoelectric (ME) effects have been studied in homogeneous systems such as single phase ceramics,[1] crystals,[2,3] and eptiaxial crystalline layers;[4] and in inhomogeneous systems such as composites of piezoelectric and magnetostrictive particles[5] and/or laminated layers.[6–16] Composites of piezoelectric $\text{Pb}(\text{Zr}_{1-x}\text{Ti}_x)\text{O}_3$ [i.e., (PZT)] ceramic or $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-x\%\,\text{PbTiO}_3$ (PMN-PT) single crystal layers laminated together with Terfenol-D,[8,9] NiFe_2O_4,[10,11] Permendur,[12] or Fe–Ga (Ref. 13) layers have been reported. The ME effect in these composites has been found to be largest for either longitudinal-transverse (LT) (Ref. 14) or longitudinal-longitudinal (LL) (Ref. 15) vibration modes.

Nearly all prior investigations have reported the ME coefficient as a voltage (ΔV_{ME}) induced by a unit applied ac magnetic field (H_{ac}), given as $\alpha_{\text{ME}}=\Delta V_{\text{ME}}/\Delta t H_{\text{ac}}$ (V/Oe cm), where α_{ME} is the ME voltage coefficient and Δt is the sample thickness. However, this coefficient is not the fundamental ME parameter, rather it is the voltage induced across an insulating ME material by H_{ac}. Early investigations[17] properly identified the fundamental parameter as the ME susceptibility (α_{me}), given as $\alpha_{\text{me}}=\delta P/\delta H_{\text{ac}}$ (in s/m), where P is the polarization. In the case that the external electric field is $E=0$, we get another formula $\alpha_{\text{me}}=\delta D/\delta H_{\text{ac}}$ (in s/m) for ME susceptibility, where D is the dielectric displacement. Early room temperature ME measurements of Cr_2O_3 crystals[2,3] reported a value of $\alpha_{\text{me}}=2.67\times10^{-12}$ s/m, which remains to this day the highest value for any single phase material. However, subsequent measurements have ignored the study of α_{me}, and rather focused on α_{ME},—which is unfortunate, as α_{me} is the fundamental ME parameter (analogous to dielectric constant and magnetic permeability), whose understanding is essential to the design devices such as gyrators[18] and E-field tunable microwave dielectric resonators.[19,20] In this letter, we will report investigations of the ME susceptibility for various ME composites, which will reveal that the fundamental parameter α_{me} is on the order of 10^{-7} s/m, which is $\sim10^5$ times larger than that of Cr_2O_3 crystals.

The ME susceptibility must be a complex quantity, similar to the effective dielectric permittivity (K_{eff}) and effective magnetic permeability (μ_{eff}) even though it has never been treated as such. Accordingly, the basic properties of the complex ME susceptibility (α_{me}^*) are as follows: (i) it is described as

$$\alpha_{\text{me}}^* = \alpha'_{\text{me}} - i\alpha''_{\text{me}} = |\alpha_{\text{me}}|e^{-i\phi}, \tag{1a}$$

where α'_{me} and α''_{me} are the real and imaginary components of the response and ϕ is the phase angle; (ii) it is connected in a general case to the ME voltage coefficient as

$$\alpha_{\text{me}}^* = |\alpha_{\text{ME}}^*|\cdot|K_{\text{eff}}^*|e^{-i\phi} = \alpha'_{\text{me}} - i\alpha''_{\text{me}}, \tag{1b}$$

where $\alpha_{\text{ME}}^*=|\alpha_{\text{ME}}|e^{-i\phi_1}$, $K_{\text{eff}}^*=|K_{\text{eff}}|e^{-i\phi 2}$, and $\phi=\phi_1+\phi_2$; and (iii) there is an inherent limit of the upper magnitude of the modulus given as

$$|\alpha_{\text{me}}| \leqslant (|K_{\text{eff}}|\cdot|\mu_{\text{eff}}|)^{1/2}, \tag{1c}$$

which is fixed by the laws of electromagnetism due to the restriction that $|\alpha|\cdot|\nu|\leqslant 1$, where ν is the speed of the electromagnetic wave in the ME media.

We measured both modulus and phase of α_{me}^* for a number of laminate composites of different configurations: (i) a longitudinally poled piezoelectric layer laminated between longitudinally magnetized magnetostrictive ones, or LL mode,[15] and (ii) a transversely poled piezoelectric layer laminated between two longitudinally magnetized ones, or LT mode.[14] Laminates of different material couples were studied, including (i) piezoelectric $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-\text{PbTiO}_3$ (PMN-PT) and magnetostrictive Terfenol-D, (ii) piezoelectric $\text{Pb}(\text{Zr}_x\text{Ti}_{1-x})\text{O}_3$ (PZT) and Terfenol-D, and (iii) PZT and magnetostrictive NiFe_2O_4 (NFO). A summary of material couples used in the hybrid and laminate configurations is given in Table I, alongside the modulus of α_{me}^* at 1 kHz. During the ME measurement, dc (H_{dc}) and ac (H_{ac}) magnetic fields were applied along the length of the laminates. An electromagnet was used to provide H_{dc}, and a Helmholtz coil

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TABLE I. Modules of the ME susceptibility for various ME laminate composites and different mode configurations.

Composite type	Mode (thickness)	α_{me} (s/m)
PMN-PT/Terfenol-*D*	LL (1 mm)	8.7×10^{-8}
	LT(0.6 mm)	5.2×10^{-8}
PZT/Terfenol-*D*	LL (2 mm)	4.2×10^{-8}
	LT (2 mm)	1.5×10^{-8}
PZT/NFO	LT(1.2 mm)	1.1×10^{-9}

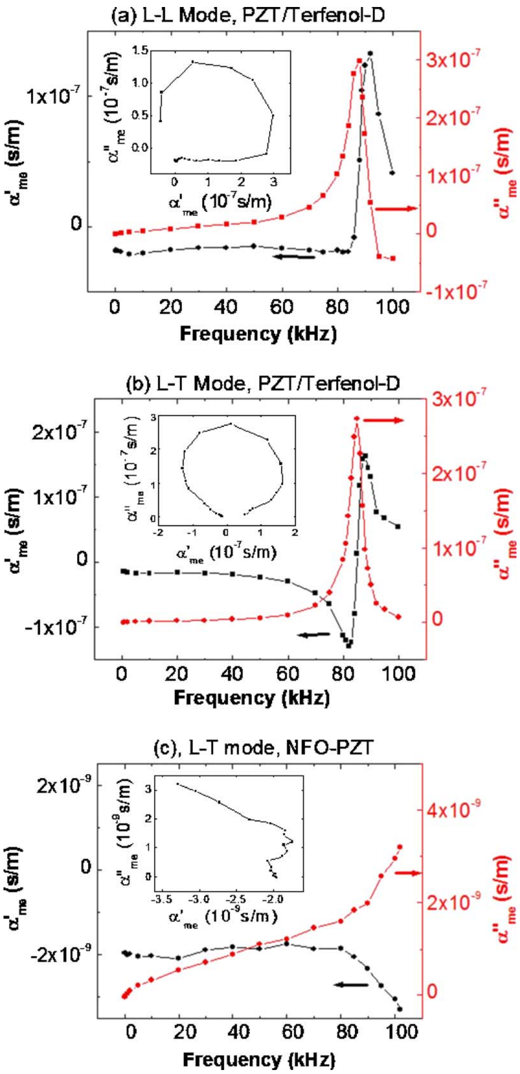

FIG. 1. (Color online) The real (left-hand side) and imaginary (right-hand side) components of the ME susceptibility (α_{me}) as a function of frequency for (a) a LL mode configuration consisting of PZT layers laminated together with Terfenol-D ones; (b) a LT configuration consisting of PZT layers laminated together with Terfenol-D ones; and (c) a LT configuration consisting of PZT layers laminated together with $\mathrm{NiFe_2O_4}$ ones. The insets show Cole-Cole or Argand-type plots of the real and imaginary components. These measurements were performed under an ac magnetic field of $H_{\mathrm{ac}}=1$ Oe and dc magnetic bias of $H_{\mathrm{dc}}=500$ Oe.

was used to generate $H_{\mathrm{ac}}=1$ Oe. A lock-in amplifier (SR850) generated a controllable input current to the Helmholtz coil, and subsequently measured the output voltage and phase from the piezoelectric layer. The complex ME susceptibility was calculated using measurements made by a lock-in method via the ME output charge and the phase shift between this output charge and input reference current. Due to a phase delay introduced by the input charge amplifier to the lock-in, we found that the phase shift between the *H*-induced voltage across the PZT and the reference current was different from that of the induced charge and reference. This was mitigated by placing large capacitors in series with the charge amplifier.

Figure 1 shows the complex ME susceptibility as a function of frequency over the bandwidth of $10 < f < 10^5$ Hz for three layer laminates operated in (a) the LL mode, consisting of Terfenol-D/PZT/Terfenol-D and (b) the LT mode consisting of Terfenol-D/PZT/Terfenol-D. In both figures, the real component α'_{me} of the ME susceptibility is given on the left-hand axis, and the imaginary one α''_{me} on the right-hand axis. The value of α^*_{me} modulus was found to be dramatically enhanced near the electromechanical resonance (EMR) frequency, similar to prior investigations of α_{ME}, reaching values as high as 3×10^{-7} s/m in both modes. At lower frequencies of $f \approx 10^3$ Hz, the modulus of α^*_{me} was found to be on the order of 10^{-8} s/m (see Table I). The imaginary component of the ME susceptibility was also found to demonstrate a strong enhancement at the EMR. Figure 1(c) shows the complex ME susceptibility as a function of frequency for NFO/PZT/NFO three layer laminates operated in the LT mode. The frequency range investigated ($10 < f < 10^5$ Hz) was insufficient to drive the laminate through its natural resonance. However, the results in the figure clearly show similar frequency dispersion of α''_{me} and α'_{me} as that given in Fig. 1.

The insets of Fig. 1 show plots of α''_{me} vs α'_{me}, known as Cole-Cole plots or Argand diagrams in dielectric or magnetic dispersion analysis, respectively. The plots of α''_{me} vs α'_{me} nearly make a complete circle when all the data pairs for the various frequencies are included over the bandwidth about the EMR. Semicircles in such plots are typical of a complete relaxing out of a contribution to a storage modulus and the loss of an underlying susceptibility mechanism. For the ME laminates, this is the loss of the elastic interaction between magnetostrictive and piezoelectric layers, as at frequencies greater than the EMR the laminates become elastically clamped. The full circle simply reflects a 180° phase shift in α''_{me} in going through the EMR, creating two semicircles.

These results clearly demonstrate that the high ME susceptibility of ME laminates is due to elastic interactions between strictions of the individual layers, and that dispersion is a natural consequence of the loss of the underlying ME mechanism as the inertial mass clamps out the samples ability to vibrate with increasing frequency. The findings firmly establish that the ME susceptibility (i) is a complex quantity, (ii) that follows conventional types of dispersion relationships, and (iii) which has enormous storage compliance of 3×10^{-7} s/m at the EMR, nearly five orders of magnitude higher than that of $\mathrm{Cr_2O_3}$ (Refs. 2 and 3), which has the highest value of any known single phase material.

We next show in Fig. 2(a) that the modulus of the ME susceptibility is connected to the modulus of the ME voltage

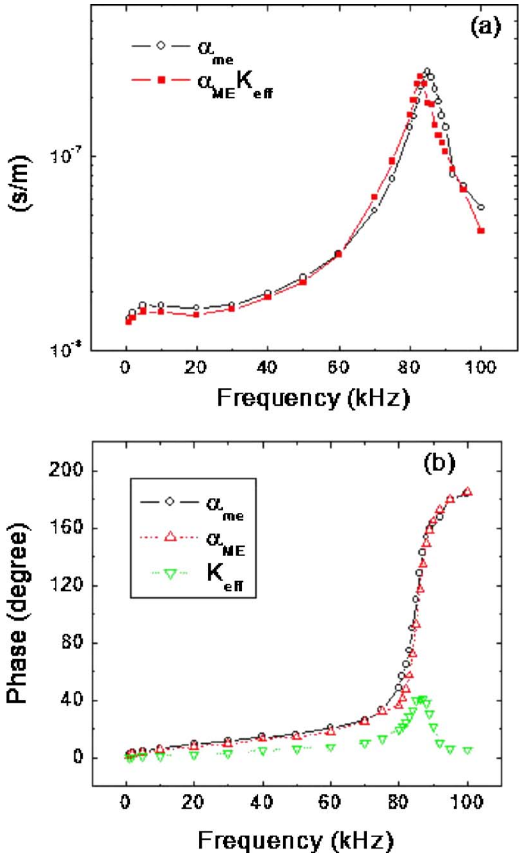

FIG. 2. (Color online) Demonstration showing the inter-relationship [fulfilling Eq. (1b)] between the ME susceptibility (α_{me}) and ME voltage coefficient (α_{ME}) for a LT PZT/Terfenol-D laminate: (a) the real components of the responses and (b) the imaginary components.

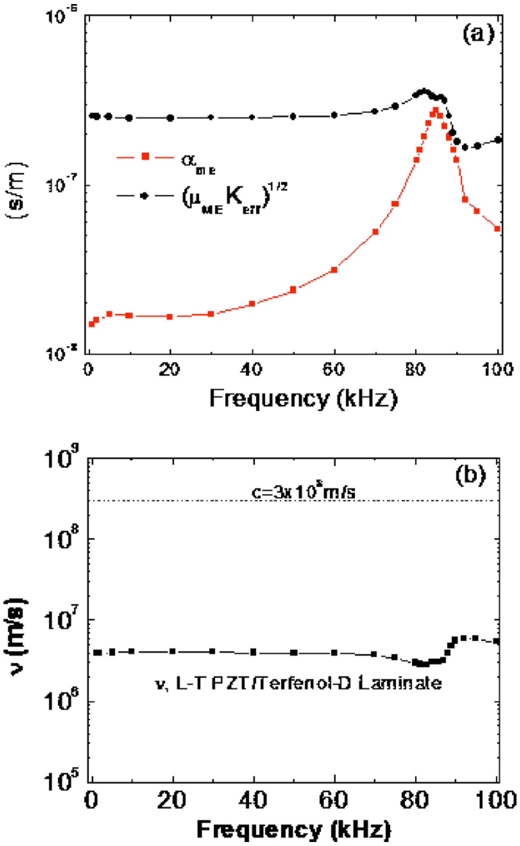

FIG. 3. (Color online) Demonstration of the inherent limit of the upper magnitude of the ME susceptibility modulus ($|\alpha_{me}|$): (a) $|\alpha_{me}|$ and $(|K_{eff}|\cdot|\mu_{eff}|)^{1/2}$, and (b) the speed of an electromagnetic wave ν in the ME media.

coefficient. These data were taken for the LT mode of a PZT/Tefenol-D laminate. In this figure, we can see that α_{me} and $\alpha_{ME}\varepsilon_{eff}$ are equivalent over the entire bandwidth range of $10<f<10^5$ Hz. Next, in Fig. 2(b), we show that the corresponding phase angle of the ME susceptibility for this same laminate is related to that of the ME voltage coefficient. Together these results show that the modulus and phase angle of the ME susceptibility are related to those of the ME voltage coefficient, as given by Eq. (1b).

Finally, we show in Fig. 3(a) the frequency dependence of the α^*_{me} modulus plotted next to that of $(|K_{eff}|\cdot|\mu_{eff}|)^{1/2}$. Inspection of this figure will reveal that Eq. (1c) is fulfilled over the entire bandwidth, i.e., $|\alpha_{me}|\leqslant(|K_{eff}|\cdot|\mu_{eff}|)^{1/2}$. We can then calculate the speed of electromagnetic radiation in the ME laminate using the fact that $|\alpha|\cdot|\nu|\leqslant 1$. We show the results for this calculation in Fig. 3(b), which gives ν as a function of the H_{ac} applied to the laminate. In this figure, it can be seen (i) that the speed of electromagnetic radiation in a ME laminate under resonant drive is a mere 3×10^6 m/s, 1% of that of electromagnetic wave in vacuum; (ii) that the modulus of $\alpha^*_{me}\approx\nu$ only in the vicinity of the EMR, and at subresonant frequencies the modulus of $\alpha^*_{me}\ll\nu$; (iii) that ν is nearly independent of the frequency of H_{ac} for that less than the EMR; and (iv) with increasing frequency and relaxing out of the elastic contribution to the ME susceptibility, the value of ν increases. These results also bring to focus the difficulty of achieving large values of α_{me}, as it requires a dramatic slowing down of ν: an instability in the modulus of α^*_{me} would require the trapping of electromagnetic radiation. Clearly, the ME susceptibility is a fundamental parameter that relates the coupling between polarization and magnetization via the media's interaction with an electromagnetic wave.

In summary, we report investigations of the complex ME susceptibility for various ME laminated composites, demonstrating that (i) the ME susceptibility is a complex quantity; (ii) the modulus of α^*_{me} is on the order of 10^{-7} s/m for various laminate configurations, and follows conventional dispersion relations as the EMR range is approached; and (iii) the velocity of electromagnetic radiation is dramatically slowed down on propagating in a media with a high α_{me}, such as a ME laminate. Our results firmly demonstrate that the ME susceptibility is a fundamental parameter: one relating the coupling of magnetization and polarization via a media's interaction with an electromagnetic radiation.

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