Resonant magnetoelectric coupling in trilayers of ferromagnetic alloys and piezoelectric lead zirconate titanate: The influence of bias magnetic field

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We present the first data and theory for the bias magnetic field dependence of magnetoelectric coupling in the electromechanical resonance (EMR) region for ferromagnetic-piezoelectric heterostructures. Trilayers of Permendur, a Co-Fe-V alloy, and lead zirconate titanate were studied. Measurements of the magnetoelectric (ME) voltage coefficient α_E indicate a strong ME coupling in the low-frequency range and a giant ME effect due to EMR at 200–300 kHz for radial modes and at ~2.7 MHz for thickness modes. Data were obtained for the bias field H dependence of two key parameters, the EMR frequency f_r and the ME coefficient $\alpha_{E,R}$ at resonance. With increasing H, an increase in f_r and a rapid rise and fall in $\alpha_{E,R}$ are measured. In our model we consider two mechanisms for the magnetic field influence on ME interactions: (i) a shift in the EMR frequency due to changes in compliance coefficients (ΔE effect) and (ii) variation in the piezomagnetic coefficient that manifests as a change in $\alpha_{E,R}$. Theoretical profiles of α_E vs frequency and estimates of frequency shift based on the ΔE effect are in excellent agreement with the data.

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I. INTRODUCTION

Magnetoelectric (ME) effects in materials are of fundamental and technological importance.[1] A composite consisting of magnetostrictive and piezoelectric phases is expected to show ME effects that are mediated by mechanical stress.[1–3] Van den Boomgaard first synthesized bulk ME composites of cobalt or nickel ferrite and $BaTiO_3$.[2,3] A multilayer structure of magnetostrictive and piezoelectric phases is predicted to a show much stronger ME coupling than bulk composites due to negligible leakage currents and a high degree of polarization.[4] Several studies in recent years reported the observation of strong ME interactions in layered systems with ferrites, manganites, or terfenol for the ferromagnetic phase and barium titanate, lead magnesium niobate-lead titanate, or lead zirconate titanate (PZT) for the piezoelectric phase.[4–11]

This work is devoted to investigations of resonant ME effects in layered systems. Studies are important at two types of resonances: ferromagnetic resonance (FMR) for the magnetic phase and electromechanical resonance (EMR) for the piezoelectric component.[12,13] For FMR studies at frequencies on the order of 9–10 GHz, the layered sample is first driven to the resonance condition and then subjected to an external electric field E' (we use the symbol E' for electric field and E for Young's modulus). The stress produced due to the piezoelectric effect at the phase boundary results in a shift δH_r in the resonance field. The parameter $A = \delta H_r / E'$ is a measure of the strength of ME coupling.[12]

Here we are concerned with the resonance phenomenon of the second kind: ME effects at EMR due to radial and thickness modes in the composite. The ME coupling at EMR is similar in nature to the low-frequency coupling—i.e., an induced polarization δP under the action of an ac magnetic field δH. But the ac field δH is tuned to EMR. As the dynamic magnetostriction is responsible for the electromagnetic coupling, EMR leads to significant increasing in the ME voltage coefficients.[13]

We provide here the first data on magnetic field effects on ME coupling at EMR and a theory for the phenomenon. A shift in the resonance frequency is expected in an applied magnetic field due to changes in Young's modulus, called the ΔE effect. Variations in the piezomagnetic coupling with H will also affect the strength of the ME coefficient at resonance. The study has been performed on a specific layered system consisting of Permendur, an alloy of Co-Fe-V with high magnetostriction and strong piezomagnetic coupling, and PZT. Trilayers were made by bonding PZT and the alloy disks. The ME voltage coefficient $\alpha_E = \delta E' / \delta H$ was determined by measuring the electric field $\delta E'$ generated across the sample in an ac field δH (from 10 Hz to 3 MHz) and a bias magnetic field H. We observed resonances in α_E versus frequency for *both* radial and thickness modes, from 200 kHz to 2.7 MHz. The resonance frequency f_r and the peak α_E were then measured as a function of H. A general increase in f_r is observed with increasing H. Theoretical estimates of the shift δf_r and peak α_E are in excellent agreement with the data. We show that $\delta f_r / H$ is a good indicator of the strength of ME coupling in the system. Details of these measurements, results, and analysis are provided in the following sections.

II. EXPERIMENT

Trilayer structures of ferromagnetic Permendur and piezoelectric PZT were synthesized. Permendur (PE) is a soft magnetic alloy consisting of 49% Fe, 49% Co, and 2% V. It is an ideal material for studies of ME composites due to a desirable low resistivity and high Curie temperature (1213 K) and high magnetostriction (70 ppm).[14] Lead zirconate titanate was chosen due to the high ferroelectric Curie temperature and piezoelectric coupling constant. Disks of PZT and PE, 9 mm in diameter and 0.18–0.8 mm in thickness, were used to make the trilayers. Samples of PZT were first poled by heating to 425 K and cooling back to room temperature in an electric field of 30–50 kV/cm perpendicular to the sample plane. Trilayers were then made with the central PZT layer bonded to outer PE layers using 0.01–0.03-mm-thick layer of a quick-dry epoxy. The thickness of the epoxy layer was found to affect the ME coupling, and the optimum thickness for maximum ME coupling was in the range 0.01–0.02 mm. The data provided here are for trilayers with a 0.02-mm-thick layer of epoxy.

For ME characterization, we measured the electric field produced by an alternating magnetic field applied to the biased composite. The samples were positioned in a measurement cell and placed between the pole pieces of an electromagnet that was used to apply the bias magnetic field H. The measurement cell consisted of a sample seat located at the center of 5-cm-diam Helmholtz coils. A 1-V signal applied to the coils produced an ac magnetic field $\delta H = 0.004-15$ Oe from 10 Hz to 3 MHz. The voltage δV across the sample was amplified and measured with an oscilloscope or a lock-in amplifier. The ME voltage coefficient is estimated from $\alpha_E = \delta E'/\delta H = \delta V/t\,\delta H$ where t is the thickness of PZT. The measurements were done for two different field orientations. With the sample plane defined by (1,2), the transverse coefficient $\alpha_{E,31}$ was measured for the magnetic fields H and δH along direction 1 (parallel to the sample plane) and perpendicular to $\delta E'$ (direction 3). The longitudinal coefficient $\alpha_{E,33}$ was measured for all the fields perpendicular to the sample plane. Magnetoelectric characterization was carried out at room temperature as a function of H and frequency of the ac magnetic field δH. For studies on ME coupling at electromechanical resonance, a high-precision signal source (Rhode and Schwarz, model SMT03) with a frequency resolution of 0.1 ppm was used.

III. RESULTS

We are primarily interested in ME coupling at EMR in the layered samples. For such studies, one first needs α_E vs H data at low frequencies for information on the optimum bias field range for strong ME interactions. This is followed by studies of the frequency dependence of α_E that shows resonance behavior for radial and thickness modes. Finally investigations of the effect of a bias magnetic field on electromechanical resonance are presented.

Low-frequency measurements of α_E vs H were carried out for in-plane (transverse) and out-of-plane (longitudinal) magnetic fields at 1 kHz. Figure 1 shows representative data

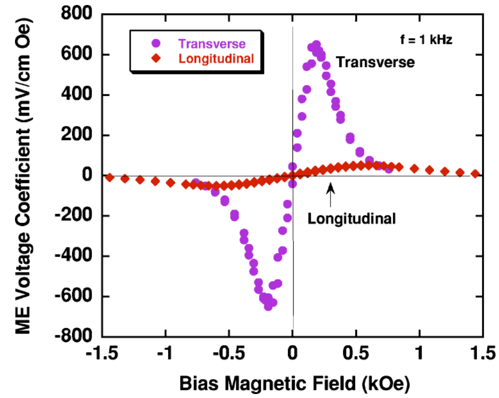

FIG. 1. (Color online) Magnetoelectric (ME) voltage coefficient $\alpha_E = \delta E'/\delta H$ versus bias magnetic field H for a Permendur (PE)—lead zirconate titanate (PZT)—PE trilayer consisting of 0.18-mm-thick layers of PE and 0.36-mm-thick layer of PZT. The data at room temperature and 1 kHz are for transverse (out-of-plane $\delta E'$ perpendicular to in-plane δH and H) and longitudinal (out-of-plane $\delta E'$, H, and δH) field orientations.

for PE-PZT-PE with 0.18-mm-thick Permendur and 0.36-mm-thick PZT, a trilayer with equal volume of the two phases. Consider first the data for the longitudinal fields. As H is increased from zero, one observes an increase in $\alpha_{E,33}$ to a maximum at $H_m = 600$ Oe. With further increase in H, $\alpha_{E,33}$ decreases rapidly to a minimum. For transverse fields, Fig. 1 shows a similar variation in the ME voltage coefficient with H as for the longitudinal case, but the maximum in $\alpha_{E,31}$ occurs at a much smaller H_m of 150 Oe. The peak value of $\alpha_{E,31} = 10\alpha_{E,33}$. Similar measurements were done on samples with a series of PZT thickness so that the volume fraction v of PZT varied from 35% to 70%. The data showed a maximum in α_E for equal volume of PE and PZT.

The key observation in Fig. 1 is the strong ME coupling with a peak α_E of 640 mV/cm Oe for transverse fields. The magnitude and field dependence of α_E in Fig. 1 are related to the variation in the piezomagnetic coupling q with H. The ME coefficients are directly proportional to $q \sim \delta\lambda/\delta H$, where λ is the magnetostriction and the H dependence tracks the slope of λ vs H. Saturation of λ at high field leads to $\alpha_E = 0$. The coefficient for longitudinal fields is quite small due to the demagnetization associated with out-of-plane magnetic fields.

The ME coefficients in Fig. 1 are one of the highest reported for two-phase composites. Systems of interest in the past were bulk samples of ferrites with barium titanate or PZT. The α_E in Fig. 1 are higher than values reported for bulk composites of cobalt ferrite or nickel ferrite (NFO) with PZT or $BaTiO_3$.[2,3] Layered composites studied so far include ferrite-PZT, lanthanum manganite-PZT, and terfenol-PZT.[4–11] For comparison, the highest value for $\alpha_{E,31}$ is 60 mV/cm Oe in bilayers of lanthanum manganites-PZT, 1500 mV/cm Oe for NFO-PZT, and 4680 mV/cm Oe for terfenol-PZT.[4–11]

Next we measured the frequency dependence of the ME coupling. For these studies for transverse fields, the bias field H was set at $H_m = 150$ Oe and $\alpha_{E,31}$ was measured as the frequency of ac field was varied from 10 Hz to 3 MHz.

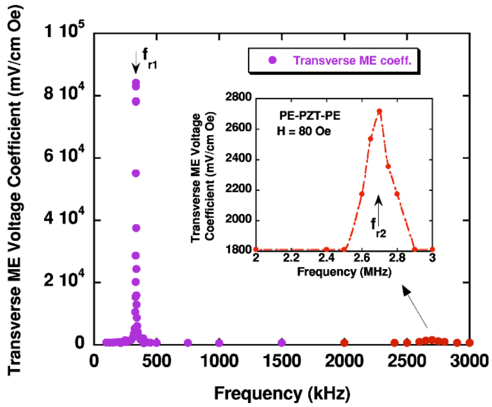

FIG. 2. (Color online) Frequency dependence of the transverse ME voltage coefficient for the PE-PZT-PE trilayer. The bias field H was set for maximum ME coupling (Fig. 1). The peaks in α_E occur at electromechanical resonance (EMR) for radial (f_{r1}) and thickness modes (f_{r2}) for the composite. The dashed line in the inset is a guide to the eye.

Typical $\alpha_{E,31}$ vs f profiles are shown in Fig. 2 for a trilayer with 0.4-mm-thick Permendur and a PZT thickness of 0.4 mm. Upon increasing f, $\alpha_{E,31}$ remains at 800 mV/cm Oe for frequencies up to 250 kHz. At higher f, we observe a rapid increase in $\alpha_{E,31}$ to a maximum of 84 V/cm Oe at 330 kHz. With further increase in f, second and third harmonics of the resonance (not shown in Fig. 2) were observed at 680 kHz and 1 MHz. Finally, $\alpha_{E,31}$ levels off at 1800 mV/cm Oe until 2.4 MHz where we see the onset of a second resonance with a peak centered at 2.7 MHz. The profile thus shows two fundamental modes: the first at $f_{1r}=330$ kHz, a half-width $\Delta f=2$ kHz, corresponding to a quality factor $Q=165$; and the second at $f_{2r}=2.7$ MHz and a Q of 14. Similar resonances were seen in $\alpha_{E,33}$ vs f profiles for longitudinal fields. The resonance occurred at the same frequency as for the transverse fields, but with a much smaller peak α_E and Q compared to the transverse fields.

The ME coefficients at resonance in Fig. 2 are a factor of 150 higher at f_{r1} and a factor 4 higher at f_{r2} compared to low-frequency values. The maximum α_E are comparable to reported values of 20–80 V/cm Oe for terfenol-PZT and ferrite-PZT.[15–18] The peaks in Fig. 2 are identified with EMR due to radial and thickness modes in the trilayer. We recently developed a model for magnetoelectric interactions at EMR for a bilayer in the form of a thin disk.[13,15] The ac magnetic field induces harmonic waves in radial or thickness modes. Based on the model, one expects a sharp increase in α_E at frequencies corresponding to these modes. Thus a dramatic enhancement of ME interactions aided by dimensional resonance occurs in the layered composites. Further details of the theory and comparison with data are provided in Sec. IV.

The effect of bias field H on ME coupling at EMR was studied by obtaining ME coefficient versus frequency profiles for a series H values. Representative profiles of $\alpha_{E,31}$ are shown in Fig. 3. With increasing H, the data show an upshift in the resonance frequency and a decrease in the peak ME coefficient $\alpha_{E,R}$.

Data on variations in f_{r1} and $\alpha_{E,R}$ with H are shown in Fig. 4. The frequency f_{r1} increases from 333 kHz for

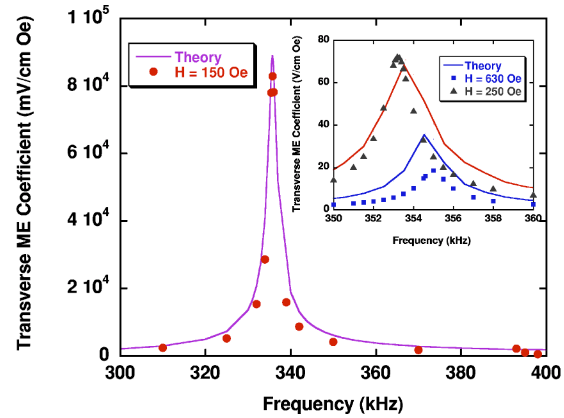

FIG. 3. (Color online) Transverse ME coefficient vs frequency profiles as in Fig. 2 for static fields of 150, 250, and 630 Oe. The lines are theoretical estimates.

$H=30$ Oe to a maximum saturation value of 336 kHz at 1 kOe. A similar measurement could not be done for the thickness mode at 2.7 MHz mainly because of a low Q for the resonance. The ME coefficient at resonance $\alpha_{E,R}$ shows a rapid increase with increasing H to a maximum of 84 V/cm Oe at $H=0.15$ kOe. A further increase in H leads to a sharp decrease in the ME coefficient. We identified two possible causes for these effects: (i) variations in the compliance coefficients in external magnetic fields (ΔE effect) and (ii) dependence of piezomagnetic coefficients on H. Further details are provided in the following section.

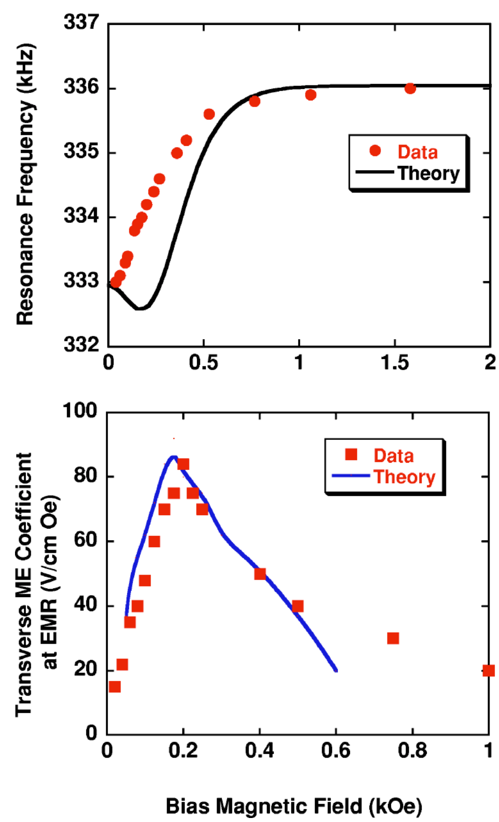

FIG. 4. (Color online) The static magnetic field dependence of the electromechanical resonance (EMR) frequency f_{r1} and ME coefficient at f_{r1}. The lines are theoretical values.

IV. DISCUSSION

The results in Sec. III provide clear evidence for strong electromagnetic coupling facilitated by mechanical stress in Permendur-PZT. We recently developed a model for low-frequency ME coupling in a bilayer.[19] The composite was considered as a homogeneous medium with piezoelectric and magnetostrictive subsystems. Expressions, in terms of the effective composite parameter, were obtained for longitudinal and transverse ME coefficients. The magnitude and H dependence of the ME voltage coefficients estimated from known material parameters for PZT and Permendur (piezoelectric coupling, magnetostriction, elastic constants, etc.) were found to be in good agreement with the data in Fig. 1.[18]

The frequency dependence of α_E in Fig. 2 that reveals peaks due to dimensional resonance is discussed next. We proposed models for ME interactions at radial and thickness modes in the layered structures.[13,15] Both radial and thickness modes were considered in a bilayer with a radius R and thickness t. The electrodes are assumed to be of negligible thickness. The composite was assumed to be a homogeneous medium that can be described by effective parameters, such as compliance, piezoelectric, and magnetostrictive coefficients that are determined from parameters for individual phases. The assumption is valid when the layer thickness is small compared to wavelengths for the acoustic modes and is certainly true for EMR that occurs at 100–3000 kHz. Based on the model, one expects resonance in α_E at frequencies that depend on R, t, compliances s_{11} and s_{12}, density and effective piezomagnetic and piezoelectric coefficients, permittivity, and loss factor.[15] Expression for the transverse ME coefficient for EMR under radial and thickness modes are given in Refs. 13 and 15. We then applied the theory to calculate $\alpha_{E,31}$ vs f for a PZT volume fraction of 0.33 in PE-PZT-PE and for the composite parameters

$$\text{PE: } s_{11} = 9 \times 10^{-12}\ \text{m}^2/\text{N}, \quad s_{12} = -3.3 \times 10^{-12}\ \text{m}^2/\text{N},$$

$$\text{PZT: } s_{11} = 15 \times 10^{-12}\ \text{m}^2/\text{N}, \quad s_{12} = -5 \times 10^{-12}\ \text{m}^2/\text{N},$$

$$\varepsilon/\varepsilon_0 = 1750, \quad d_{13} = -125 \times 10^{-12}\ \text{m/V}.$$

Apart from these parameters, we also require knowledge of the piezomagnetic coupling $q = \delta\lambda/\delta H$, which can be estimated from the variation of the magnetostriction λ with H. We measured the magnetostriction in PE-PZT-PE with the standard strain gauge technique and obtained data on λ_{11} (in-plane parallel magnetostriction) and λ_{12} (in-plane perpendicular magnetostriction). The piezomagnetic coupling coefficient for transverse ME coupling $q = q_{11} + q_{12}$ was then determined from these data. Estimates of q vs H are shown in Fig. 5 for PE-PZT-PE.

Theoretical $\alpha_{E,31}$ vs f profiles were determined using appropriate expressions from Ref. 13, the above material parameters for PE and PZT, and the q value in Fig. 5. Figure 3 shows the profile for H=150, 250, and 630 Oe along with data for comparison. There is very good agreement between theory and the data in Fig. 3.

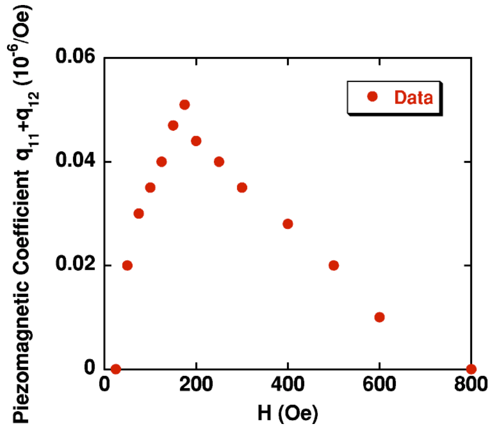

FIG. 5. (Color online) Piezomagnetic coefficient versus H data for PE-PZT-PE.

The primary focus of this work is on the effects of H on ME coupling parameters at EMR, which are discussed next. The data in Figs. 3 and 4 clearly indicate a shift in the EMR frequency H. This shift is due to a change in Young's modulus (E) caused by the magnetostriction: namely, the ΔE effect. In cubic metals and alloys there are two kinds of ΔE effect. In the first kind, E increases with the sample magnetization, whereas in the second type E decreases at first and then increases to a value higher than in the demagnetized state.[20] The change ΔE is related to domain wall motion and domain rotation and is given by

$$\frac{\Delta E}{E} = \frac{E - E_0}{E}, \tag{1}$$

where E_0 is Young's modulus at H=0. It is assumed that $\Delta E \ll E, E_0$. The effect can be described by a statistical theory of ferromagnetic domains.[21,22] The free energy density of a cubic crystal in a magnetic field can be expressed as

$$\begin{aligned} F = F_0 - M_s H(\alpha_1\alpha_1' + \alpha_2\alpha_2' + \alpha_3\alpha_3') + B_1(\alpha_1^2 S_1 + \alpha_2^2 S_2 + \alpha_3^2 S_3) \\ + B_2(\alpha_1\alpha_2 S_6 + \alpha_1\alpha_3 S_5 + \alpha_2\alpha_3 S_4) + \tfrac{1}{2}c_{11}(S_1^2 + S_2^2 + S_3^2) \\ + \tfrac{1}{2}c_{44}(S_4^2 + S_5^2 + S_6^2) + c_{12}(S_1 S_2 + S_1 S_3 + S_2 S_3). \end{aligned} \tag{2}$$

Here α_1, and α_1' are directional cosines of magnetization and magnetic field, M_s is the saturation magnetization, B_1 and B_2 are the magnetoelastic constants, and c_{ij} is the stiffness coefficient.

Using statistical averaging methods one gets the following expression for the variation in Young's modulus for a ferromagnet with [100] easy direction:

$$\frac{\Delta E}{E} = \frac{2}{9} E \Lambda t_1^2 \frac{[\cosh(h) - 1][\cosh(h) - 4]}{[\cosh(h) + 2]^2}. \tag{3}$$

For a ferromagnet with [111] easy direction,

$$\frac{\Delta E}{E} = \frac{4}{27} E \Lambda t_2^2 \frac{[\cosh(h) - \cosh(h/3)][\cosh(h) - 9\cosh(h/3)]}{[\cosh(h) + 3\cosh(h/3)]^2}. \tag{4}$$

Here $h=3\chi_0 H/M_s$ is a dimensionless parameter, χ_0 is the permeability at $H=0$, $\Lambda = 3\chi/M_s^2$, and

$$t_1 = -B_1 \frac{(c_{11}+2c_{12})}{c_{11}(c_{11}+c_{12}) - 2c_{12}^2}, \quad t_2 = -B_2 \frac{1}{2c_{44}}.$$

Using Eqs. (3) and (4) one can calculate the relative variation in E and the change in the stiffness coefficients c_{11} and c_{12} so that the shift δf_r in the EMR frequency can be estimated as a function of H based on calculation results[23] as follows:

$$\delta f_r = \frac{\Delta E f_r}{2E(1+\nu)z} \frac{(3+\nu)z^2 J_0^2(z) - 8zJ_0(z)J_1(z) + [(1+\nu)z^2 + 4(1-\nu)]J_1^2(z)}{z[J_0^2(z) + J_1^2(z)] - 2J_0(z)J_1(z)}, \tag{5}$$

where $z=kR$, $\nu=-s_{12}/s_{11}$ is Poisson's ratio, $k = \sqrt{\rho s_{11}(1-\nu^2)}\omega$, ρ is the density, ω is the angular frequency, and $J_0(k)$ and $J_1(k)$ are Bessel functions of the first kind. We used stiffness coefficients of (in units of 10^{12} N/m^2) $c_{11} = 0.19$ and $c_{12}=0.11$ for Permendur. Calculated δf_r according to Eq. (5) and the data are compared in Fig. 4. There is excellent agreement between theory[13] and data.

The bias magnetic field dependence of peak ME voltage coefficient at EMR in Figs. 3 and 4 may be described by[23]

$$\alpha_{E,R} = \frac{1}{\Gamma}\left[\frac{d_{31}(q_{11}+q_{12})}{\varepsilon_{33}s_{11}(1-\nu)}\left(\frac{(1+\nu)J_1(\kappa)}{\Delta_r} - 1\right) + \frac{m_{31}}{\varepsilon_{33}}\right], \tag{6}$$

where q_{ij} and d_{ij} are piezomagnetic and piezoelectric coefficients, ε_{ij} is the permittivity matrix, m_{33} is the ME susceptibility, $\Delta_r = \kappa J_0(\kappa) - (1-\nu)J_1(\kappa)$, and Γ is the loss factor (in our case $\Gamma=0.08$). Since $\alpha_{E,R}$ depends only on one H-dependent term—i.e., piezomagnetic coefficients—Eq. (5) may be simplified as

$$\alpha_{E,R} = g(q_{11}+q_{12}), \tag{7}$$

where g is a coefficient that is determined by mechanical, piezoelectric, geometric, and permittivity parameters of composite phases. Data on the piezomagnetic coefficient $q=q_{11}+q_{12}$ for the composite are shown in Fig. 5. These coefficients were used for theoretical values of $\alpha_{E,R}$ as a function of H. Estimated values of $\alpha_{E,R}$ using Eq. (7), q in Fig. 5, and $g=2\times10^9$ V/cm are plotted in Fig. 4. An excellent agreement between theory and data is evident in the figure.

For ME coupling at ferromagnetic resonance, we define $\delta H_r/E'$ as a measure of the strength of the interactions, where δH_r is the shift in the resonance field produced by an electric field E'. An analogous parameter for EMR is $\delta f_r/H$. Figure 6 shows the variation in $\delta f_r/H$ with H for PE-PZT-PE. The figure also shows, for comparison, data on the strength of the low-frequency ME voltage coefficient α_{E31} vs H. It is remarkable that both low- and high-frequency couplings track each other as H is increased. These observations reinforce the basic fact that the Joule magnetostriction due to domain wall motion and domain rotation is the cause of dynamic magnetoelectric interactions at low frequencies and variations in ME parameters at EMR. The H-induced changes in resonance frequency and the coupling strength are quite substantial and the technique can be utilized to study the nature of the ΔE effect in two-phase-layered ME composites.

V. CONCLUSION

This study constitutes the first report on the nature of the bias magnetic field dependence of resonance magnetoelectric interactions in trilayers of a ferromagnetic alloy and PZT. The samples contained Permendur for the ferromagnetic phase. Data on bias magnetic field dependence of ME voltage coefficients at low frequencies reveal strong ME coupling; the magnitude of α_E scales with the strength of piezomagnetic coefficient. High-frequency ME interactions have been investigated through the frequency dependence of α_E and the effect of H on electromechanical resonance. Data on α_E vs f show resonant characteristics corresponding to radial and thickness modes in the samples. A two-to-three orders of

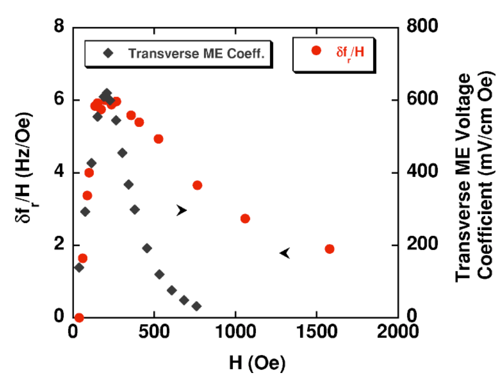

FIG. 6. (Color online) Comparison of the strength ME coupling at EMR, $\delta f/H$, and low-frequency transverse coupling coefficient $\alpha_{E,31}$ for PE-PZT-PE.

magnitude increase in α_E compared to low-frequency values is observed at EMR. Our model accounts for the observed frequency dependence of α_E. A general increase in the resonance frequency with increasing H is measured. Theoretical predictions based on the ΔE effect in the ferromagnetic metal are in excellent agreement with the data on the H dependence of the resonance frequency and ME coefficient at resonance.

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