Magnetic and magnetoelectric susceptibilities of a ferroelectric/ferromagnetic composite at microwave frequencies

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A phenomenological theory of magnetic and magnetoelectric (ME) susceptibilities of ferroelectric/ferromagnetic composites is presented and applied to the special case of layered structures. Expressions have been obtained relating the magnetic and ME susceptibility tensor components of the composite (symmetry point group $3m$ and $4mm$) to ME coupling constants. The theory predicts a unique resonance in the *electric-field dependence* of the magnetic susceptibility. It is shown that the ME susceptibility is the product of magnetic susceptibility, composite magnetization, and ME coupling constants. The model is used to obtain the magnetic and ME susceptibilities versus electric-field profiles for three bilayer composites of importance: lithium ferrite (LFO)–lead zirconate titanate (PZT), nickel ferrite (NFO)-PZT, and yttrium iron garnet (YIG)-PZT. Our calculations reveal the largest electric-field effects for NFO-PZT and the weakest effect for YIG-PZT. Three measurement methods for ME susceptibility, resonant ME coupling, electrical dipole transitions, and ME effect at ferromagnetic resonance (FMR) are proposed. As an example, we consider multilayers of LFO-PZT and determine the ME constants from data on electric-field influence on FMR. The ME parameters are then used to calculate the susceptibilities. The results indicate strong high-frequency ME effects in the composite. The theory is useful for measurements of ME susceptibility and for the design and analysis of electrically controlled high-frequency magnetic devices.

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I. INTRODUCTION

There has been considerable interest in the physics of heterostructures in recent years for possible applications in devices. Most studies were devoted to superlattices or bilayers involving the combination of ferromagnetic/nonmagnetic/antiferromagnetic layers.[1,2] Dipole and exchange interactions in magnetic superlattices are predicted to show collective spin wave and magnetostatic bulk and surface modes with unique characteristics.[1–4] Some recent theoretical studies also dealt with ferroelectric/(anti)ferroelectric bilayers.[5,6] It was shown that the hysteresis loops in such structures were dependent sensitively on the layer thickness and interface coupling. These studies suggested the possibility of multilayer ferroelectric elements for use in memory devices.[5,6]

The present work is concerned with a composite consisting of both ferromagnetic and ferroelectric phases. The focus is on magnetoelectric interactions at microwave frequencies. The magnetoelectric effect (ME) is defined as the dielectric polarization of a material in an applied magnetic field or an induced magnetization in an external electric field. The effect facilitates the conversion between energies stored in magnetic and electric fields and requires the coexistence of long-range magnetic order and ferroelectric order.[7–12] Several single-phase materials show rather weak ME effects.[10] But in a composite consisting of both magnetostrictive and piezoelectric phases, the ME effect is the result of product-property, i.e., the mechanical deformation due to magnetostriction results in a dielectric polarization due to piezoelectric effects.[7–12] Studies show strong ME effects in bulk and layered samples consisting of a ferrite that is ferrimagnetic and magnetostrictive and barium titanate or lead zirconate titanate (PZT) that is ferroelectric with strong piezoelectric coupling.

Investigations of magnetoelectric (ME) effects in composites have mainly focused on low-frequency (10–1000 Hz) effects. Harshe *et al.* proposed a model for low-frequency ME effects in ferrite–lead zirconate titanate (PZT) bilayer.[10] Other works of significance related to ME composites were reported in the MEIPIC-2 conference.[13] Getman considered the composites as a homogeneous medium and obtained theoretical concentration dependence of ME coefficients for bulk and layered samples using the effective parameters approach.[14] Lopatin *et al.* carried out experimental studies on multilayers of ferrite-PZT composites and observed strong ME effects.[15] Our very recent works on bilayers and multilayers of nickel ferrite (NFO), lithium ferrite (LFO), or lanthanum strontium manganite with PZT indicated a giant low-frequency ME effect with a coupling coefficient ranging from 60–1500 mV/cm Oe.[11,12]

The primary focus of this work is on the high frequency effects in artificial heterostructures of ferrite-PZT. Most iron based ferrimagnetic oxides show two high-frequency resonances in the magnetic susceptibility: one in the microwave region associated with the net ferromagnetic moment and a second resonance at a higher frequency corresponding to the antiferromagnetic alignment of sublattice moments. It is therefore important to study the effects of ME coupling at frequencies close to both resonance regimes. Tilley and Scott provided a theory of such high-frequency ME effects for a single phase ME material, antiferromagnetic $BaMnF_4$.[16]

They incorporated ME interactions in the free-energy expressions, and with the aid of the Landau-Lifshitz equations of motions showed that with increasing strength of ME interactions (i) the magnon frequency decreased and (ii) the optical-phonon frequency increased. It was also shown that magnetic, dielectric, and magnetoelectric susceptibilities have poles at magnon and optical-phonon frequencies.

Since ferrites are materials of choice for microwave signal processing devices due to low losses, ferrite-PZT composites are promising candidates for a class of high-frequency devices based on ME interactions. This work therefore focuses on the properties of the composite in the microwave region of the electromagnetic spectrum. In our recent work, we considered a ferrite-PZT bilayer subjected to a microwave magnetic field and constant E and H fields.[17] We developed a phenomenological theory for the shift of magnetic resonance field induced by external electric field and obtained an expression for the shift in terms of ME coupling constants. The shift occurs due to electric-field induced strain in PZT and ME coupling. Assuming nominal values for ME coupling constants, we estimated the field shift for NFO-PZT and yttrium iron garnet (YIG)-PZT layered composites as a function of the volume ratio for the two phases. Alternately, the theory enables the determination of ME constants from data on E induced field shift in the magnetic resonance region.

In this study, we address the following questions of importance on the high-frequency response of the ferrite based composites.

(1) We provide a general theory applicable to both bulk and layered samples that are subjected to dc and ac magnetic and electric fields. Here we consider two types of interactions: (i) coupling between microwave magnetic field and dc electric field, and (ii) coupling between microwave magnetic and electric fields.

(2) The former coupling is expressed in terms of electric-field effects on magnetic susceptibility. The theory predicts a resonance in the E dependence of the susceptibility.

(3) The interaction between ac electric and magnetic fields is described in terms of a ME susceptibility tensor and explicit expressions are derived for the tensor components. The theory when applied to bilayers of ferrite-PZT predicts a strong microwave ME effect in nickel ferrite-PZT.

(4) We propose three measurement techniques for ME susceptibility in the microwave region.

(5) Finally, data on ME coupling constants obtained for a multilayer ferrite-PZT samples are used to estimate the magnetic and ME susceptibilities for comparison with theory.

The study is also essential for potential use of the composites in devices. With the composite, traditional magnetic devices such as a microwave switch or a phase shifter could be controlled with an external electric field [interaction of type (i)].[18,19] There are also possibilities for devices, including magnetoelectric spin-wave amplifiers in which rf electric fields facilitate the amplification and coupled-wave devices in which hybrid spin-electromagnetic or spin-acoustic waves are produced through ME coupling [interaction of type (ii)].

In Sec. II we consider the type (i) interaction for a sample subjected to dc and ac magnetic fields. The model is used to obtain the magnetic susceptibility versus electric-field profiles in a bilayer of LFO-PZT, NFO-PZT, and YIG-PZT. For the calculations, we use theoretical values of ME coupling constants obtained from elastic constants and piezoelectric and magnetostriction parameters, as discussed in Ref. 17. Our calculations reveal the largest electric-field effects on magnetic susceptibility for NFO-PZT composites. In Sec. III we consider a composite subjected to time-dependent electric and magnetic fields [type (ii) interaction], and analyze microwave ME susceptibility.[20,21] It is shown that the ME susceptibility is the product of magnetic susceptibility, composite magnetization and ME coupling constants. We again consider LFO-PZT, NFO-PZT, and YIG-PZT bilayers and estimate ME susceptibilities vs H profiles as a function of applied electric fields, called the magnetoelectric spectrum. We observe a resonance in the ME spectrum, with NFO-PZT system showing the largest resonance amplitude. The ME susceptibility is a key parameter for the design of devices based on microwave magnetoelectric coupling. In Sec. IV we propose two direct and an indirect measurement methods for magnetoelectric susceptibility. The direct procedures involve (i) resonant coupling of microwave fields in the composite placed in a resonator or transmission line and (ii) power absorption due to electric dipole transitions. We demonstrate the usefulness of this procedure with specific reference to LFO-PZT composites.

II. INFLUENCE OF A CONSTANT ELECTRIC FIELD ON MAGNETIC SUSCEPTIBILITY

In a composite, the interaction between electric and magnetic subsystems can be expressed in terms of a ME susceptibility. In general, the susceptibility is defined by the following equations:[13,22,23]

$$\mathbf{P}=\chi^{E}\mathbf{E}+\chi^{EM}\mathbf{H},$$

$$\mathbf{M}=\chi^{ME}\mathbf{E}+\chi^{M}\mathbf{H}. \tag{1}$$

Here $\mathbf{P}$ is the electrical polarization, $\mathbf{M}$ is the magnetization, $\mathbf{E}$ and $\mathbf{H}$ are the external electrical and magnetic fields, χ^{E} and χ^{M} are the electrical and magnetic susceptibilities, and χ^{EM} and χ^{ME} are the ME susceptibilities, with $\grave{\chi}^{IE}_{ik}=\grave{\chi}^{EI}_{ki}$. In Eq. (1) $\mathbf{P}$, $\mathbf{M}$, $\mathbf{E}$, and $\mathbf{H}$ generally contain both dc and ac components, for example, for $\mathbf{P}$

$$\mathbf{P}=\mathbf{P}_{0}+\mathbf{p}\exp(i\omega t).$$

For the microwave regime, the ME susceptibility is defined by the expressions

$$\mathbf{p}=\chi^{E}\mathbf{e}+\chi^{EM}\mathbf{h},$$

$$\mathbf{m}=\chi^{ME}\mathbf{e}+\chi^{M}\mathbf{h}. \tag{2}$$

In Eq. (2), the ac amplitudes are shown explicitly, but the susceptibilities also depend on constant fields.

We consider a sample subjected to constant electric and magnetic fields and a variable magnetic field. The thermodynamic potential density can be written as

$$W=W_0+W_{\mathrm{ME}}, \tag{3}$$

where W_0 is the thermodynamic potential density at $E=0$, and

$$W_{\mathrm{ME}}=B_{ikn}E_iM_kM_n+b_{ijkn}E_iE_jM_kM_n. \tag{4}$$

Here B_{ikn} and b_{ijkn} are linear and bilinear ME constants, respectively. The number of independent components is determined by the structure of a material. The main contribution to W_{ME} in polarized composites arises from the linear ME constants B_{ikn}. If the composite is unpolarized, the bilinear ME constants are dominant. Both the linear and bilinear ME terms are important if the composite is partially polarized.[24] We used the effective demagnetization factor method to solve the linearized equation of motion of the magnetization and obtained the following expression for the magnetic susceptibility:

$$\chi^M=\begin{bmatrix}\chi_1 & \chi_s+i\chi_a & 0\\ \chi_s-i\chi_a & \chi_2 & 0\\ 0 & 0 & 0\end{bmatrix}, \tag{5}$$

where

$$\chi_1=D^{-1}\gamma^2M_0\Big[H_{03'}+M_0\sum_i (N^i_{2'2'}-N^i_{3'3'})\Big];$$

$$\chi_2=D^{-1}\gamma^2M_0\Big[H_{03'}+M_0\sum_i (N^i_{1'1'}-N^i_{3'3'})\Big];$$

$$\chi_s=-D^{-1}\gamma^2M_0^2\sum_i N^i_{1'2'};$$

$$\chi_a=D^{-1}\gamma M_0\omega;$$

$$D=\omega_0^2-\omega^2;$$

$$\omega_0^2=\gamma^2\Big[H_{03'}+\sum_i (N^i_{1'1'}-N^i_{3'3'})M_0\Big]\times\Big[H_{03'}+\sum (N^i_{2'2'}-N^i_{3'3'})M_0\Big]-\Big(\sum_i N^i_{1'2'}M_0\Big)^2.$$

Here γ is the magnetomechanical ratio, ω is the angular frequency, $N^i_{k'n'}$ are demagnetization factors describing the effective magnetic anisotropy fields, and $1',2',3'$ is a coordinate system in which the axis $3'$ is directed along the equilibrium magnetization. In Eq. (5) the summation is carried out over all types of magnetic anisotropy. The ME interaction results in an additional term ($i=$Å),

$$N^E_{k'n'}=2(B_{ikn}+b_{ijkn}E_{oj})E_{oi}\beta_{k'k}\beta_{n'n}, \tag{6}$$

where is matrix of direction cosines of axes $(1',2',3')$ relative to the crystallographic coordinate system (1, 2, 3).

We shall now derive a general expression for the magnetic susceptibility tensor of ferrite-ferroelectric composite assuming a structure with $3m$ or $4mm$ symmetry point group. Equation (4) in this case becomes

$$\begin{aligned}W_{\mathrm{ME}}={}&E_1[-2B_{22}M_1M_2+2B_{15}M_1M_3]+E_1[B_{22}(M_2^2-M_1^2)+2B_{15}M_2M_3]+E_3(B_{33}-B_{31})M_3^2+E_1^2[(b_{11}-b_{12})M_1^2\\&+(b_{13}-b_{12})M_3^2+2b_{14}M_2M_3]+E_2^2[(b_{11}-b_{12})M_2^2+(b_{13}-b_{12})M_3^2-2b_{14}M_2M_3]+E_3^2(b_{33}-b_{31})M_3^2\\&+2E_2E_3(b_{41}M_1^2-b_{41}M_2^2+2b_{44}M_2M_3)+4E_1E_3(b_{44}M_1M_3+B_{41}M_1M_2)+4E_1E_2(b_{14}M_1M_3+b_{66}M_1M_2).\end{aligned} \tag{7}$$

$b_{66}=(b_{11}-b_{12})2$ for $3m$ symmetry point group and $B_{22}=b_{14}=b_{41}=0$ for $4mm$ symmetry point group. We shall assume the matrix of the form

$$\hat{\beta}=\begin{pmatrix}1 & 0 & 0\\ 0 & \cos\Theta & -\sin\Theta\\ 0 & \sin\Theta & \cos\Theta\end{pmatrix}, \tag{8}$$

where Θ is the angle between the equilibrium magnetization and the symmetry axis of the crystal. Considering a disk sample in the basal plane and with a uniaxial magnetic anisotropy, we get from Eqs. (6), (7), and (8)

$$\begin{aligned}\sum_i (N^i_{1'1'}-N^i_{3'3'})={}&\Big(\frac{2K_1}{M_0^2}-4\pi+2q\Big)\cos^2\Theta+2r\sin 2\Theta\\&+2(b_{11}-b_{12})(E_{01}^2-E_{02}^2)\\&+8b_{41}E_{02}E_3,\end{aligned}$$

$$\sum_i (N^i_{2'2'}-N^i_{3'3'})=\Big(\frac{2K_1}{M_0}-4\pi+2q\Big)\cos 2\Theta+4r\sin 2\Theta, \tag{9}$$

$$\begin{aligned}\sum_i N^i_{1'2'}={}&4(b_{66}E_{01}E_{02}+b_{11}E_{01}E_{03})\cos\Theta\\&-4(b_{14}E_{01}E_{02}+b_{44}E_{01}E_3)\sin\Theta,\end{aligned}$$

where

$$\begin{aligned}N^E_{1'1'}-N^E_{3'3'}={}&-4B_{22}E_{02}+2(b_{11}-b_{12})(E_{01}^2-E_{02}^2)\\&+8b_{41}E_{02}E_{03}+2g_2\cos^2\Theta+2g_3\sin 2\Theta,\end{aligned}$$

$$N^E_{2'2'}-N^E_{3'3'}=2g_2\cos 2\Theta+4g_3\sin 2\Theta, \tag{10}$$

$$N^E_{1'2'}=[2B_{22}E_{01}+4(b_{41}E_{01}E_{03}+b_{66}E_{01}E_{02})]\cos\Theta$$
$$-(2B_{15}E_{01}+4b_{44}E_{01}E_{03})\sin\Theta,$$

$$g_2=B_{22}E_{02}+(B_{31}-B_{33})E_{03}+(b_{12}-b_{13})E_{01}^2+(b_{11}$$
$$-b_{13})E_{02}^2+(b_{31}-b_{33})E_{03}^2-2b_{41}E_{02}E_{03},$$

$$g_3=b_{14}(E_{02}^2-E_{01}^2)+2b_{44}E_{02}E_{03}.$$

Substituting Eq. (9) in Eq. (6) it is possible to get an expression for components of the magnetic susceptibility tensor. We consider the special case of the electric field directed along a symmetry axis of the structure, i.e., $Å_{01}=Å_{02}=0$, $Å_{03}=E_0$ and obtain

$$N^E_{1'1'}-N^E_{3'3'}=2[(B_{31}-B_{33})E_0+(b_{31}-b_{33})E_0^2]\cos^2\Theta,$$
$$N^E_{2'2'}-N^E_{3'3'}=2[(B_{31}-B_{33})E_0+(b_{31}-b_{33})E_0^2]\cos 2\Theta, \quad (11)$$
$$N^E_{1'2'}=0.$$

For a disk-shaped sample magnetized along the symmetry axis, Eqs. (5) and (11) yield

$$\chi_1=\chi_2=D^{-1}\gamma^2M_0H_{\text{eff}},$$
$$\chi_s=0, \quad (12)$$
$$\chi_a=D^{-1}\gamma M_0\omega,$$

where

$$D=\omega_0^2-\omega^2,$$
$$\omega_0=\gamma H_{\text{eff}},$$
$$H_{\text{eff}}=H_0+2H_a-4\pi M_0+2M_0(B_{31}-B_{33})E_0$$
$$+2M_0(b_{31}-b_{33})E_0^2,$$
$$H_a=K_1/M_0.$$

Assuming the dissipative term in the equation of motion of magnetization as $i\omega\alpha(\mathbf{M}_0\times\mathbf{m})/M_0$, where α is the dissipation parameter, the magnetic susceptibility tensor components are complex and take the form $\chi_1=\chi'+i\chi''$, where

$$\chi'=\chi_0\frac{\omega_0^2(\omega_0^2-\omega^2+2\alpha^2\omega^2)}{(\omega_0^2-\omega^2)^2+4\alpha^2\omega_0^2\omega^2},$$
$$\chi''=\chi_0\frac{\alpha\omega\omega_0(\omega_0^2+\omega^2)}{(\omega_0^2-\omega^2)^2+4\alpha^2\omega_0^2\omega^2},\quad \chi_0=\gamma\frac{M_0}{\omega_0}. \quad (12a)$$

It follows from Eqs. (5) and (12) that the dependence of the magnetic susceptibility on the dc electric field has a resonant character. The nature of that dependence can be explained as follows. By means of ME interactions, the external electric field results in a change in the effective magnetic field H_{eff} in Eq. (12) with

$$2H_{\text{ME}}=2M_0(B_{31}-B_{33})E_0+2M_0(b_{31}-b_{33})E_0^2.$$

The change originates from the piezoelectric phase mechanically coupled to the magnetostrictive phase, and is phenomenological described by ME constants B_{ikn} and b_{ijkn} in Eqs. (3) and (4). Thus the variation of the dc electric field has the same effect as magnetic-field variations and reveals a resonant behavior. Expressions for the susceptibility components could be obtained with the help of demagnetization factors stipulated by ME interactions according to Eq. (5).

Next we consider specific composites and estimate the magnetic susceptibility and its electric-field variation. Three composites of importance for the estimation are LFO-PZT, NFO-PZT, and YIG-PZT because of desirable high-frequency properties of LFO, NFO, and YIG. We consider a simple structure, a bilayer consisting of single ferrite and PZT layers. In order to obtain the susceptibilities, one requires the knowledge of ME constants and the loss parameter α. The ME constants could be estimated from the elastic constants, piezoelectric coupling constant, magnetostriction, and magnetization for the two phases as discussed in our earlier work [Eq. (9) in Ref. 17]. The elastic constants, piezoelectric coupling, and the magnetization for YIG-PZT and NFO-PZT used in the calculation are the same as in Ref. 17. For LFO the following elastic and magnetic parameters are used: ${}^mc_{11}=24.47\times10^{10}$ N/m^2; ${}^mc_{12}=13.71\times10^{10}$ N/m^2; ${}^mc_{44}=9.36\times10^{10}$ N/m^2; $4\pi M_s=3600$ G. Assuming that the poling axis of the piezoelectric phase coincides with the [100] axis of the magnetostrictive phase and a magnetostriction of $\lambda_{100}=1.4\times10^{-6}$, 23×10^{-6}, and 46×10^{-6} for YIG-PZT, LFO-PZT, and NFO-PZT, respectively,[25] we obtain $2M_0(B_{31}-B_{33})=0.1$, 0.6 and 1.4 Oe cm/kV for the three bilayers. Finally, these ME parameters and the loss parameter $\alpha=0.025$, 0.05, and 0.075 for YIG-PZT, LFO-PZT, and NFO-PZT, respectively,[25] were used to estimate the magnetic susceptibility.

Figure 1 shows static magnetic-field dependence of real and imaginary parts of magnetic susceptibility with and without an external electric field for layered LFO-PZT, NFO-PZT, and YIG-PZT. Both real and imaginary parts are shown as a function of H for $E=0$ and 300 kV/cm. The results are for a bilayer disk sample with the H and E fields perpendicular to the sample plane and for a frequency of 9.3 GHz. The static field range is chosen to include ferromagnetic resonance in the ferrite. For $E=0$, one observes the expected resonance in χ' and χ'' profiles. With the application of $E=300$ kV/cm, a downshift in the resonance field H_r occurs since the electric field essentially gives rise to an internal magnetic field. The magnitude of the shift δH varies from a maximum of 330 Oe for NFO-PZT to a minimum of 22 Oe for YIG-PZT. The parameter δH is determined by ME constants which in turn is strongly influenced, among other factors, by the magnetostriction. The large λ for NFO leads to a relatively strong E-induced effect in NFO-PZT compared to YIG-PZT.

Figure 2 shows the estimated variation of the real and imaginary parts of the magnetic susceptibility as a function of E for a frequency of 9.3 GHz. The constant magnetic field is set equal to H_r. Figure 2 then essentially shows the

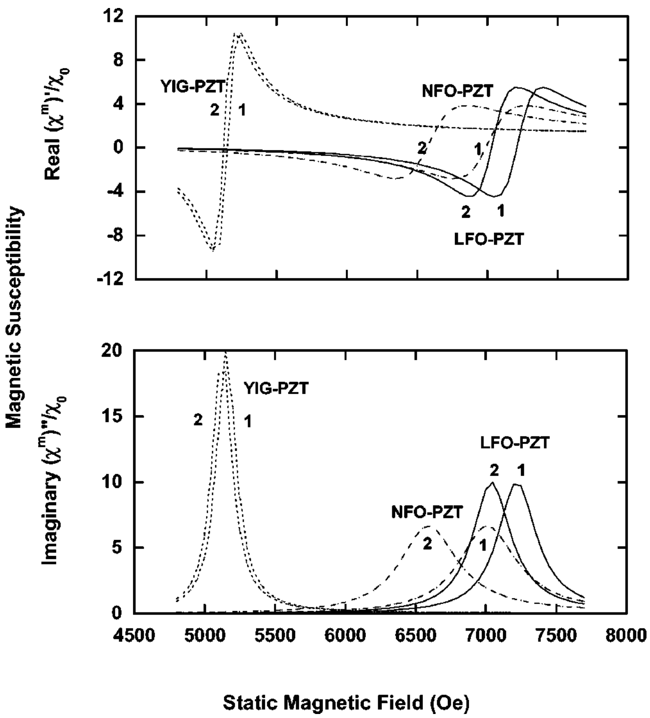

FIG. 1. Theoretical static magnetic-field dependence of the magnetic susceptibility for layered composites consisting of a single ferrite layer and a layer of lead titanate zirconate (PZT). The results are for lithium ferrite (LFO)-PZT, nickel ferrite (NFO)-PZT, and yttrium iron garnet (YIG)-PZT bilayers. The real and imaginary parts of the susceptibility at 9.3 GHz are for an applied electric field of (1) $E=0$ and (2) $E=300$ kV/cm. Notice the downshift in the resonance field when E is increased from 0 to 300 kV/cm.

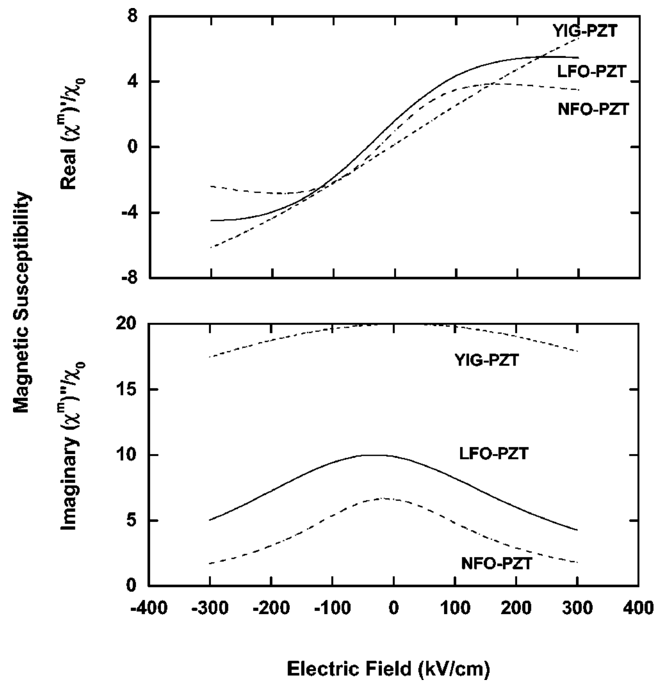

FIG. 2. Calculated electric-field dependence of the magnetic susceptibility at 9.3 GHz for bilayers of LFO-PZT, NFO-PZT, and YIG-PZT. The static magnetic field H corresponds to the resonance field for $E=0$.

electric-field dependence of the susceptibility components and the resulting resonance. One notices a linear dependence of χ' on E for YIG-PZT and the variation is nonlinear for the other two samples. The imaginary component generally decreases with increasing magnitude of E, with the largest influence for NFO-PZT. The width of the resonance in Fig. 2 measured in terms of electric field is inversely proportional to the parameter $2M_0(B_{31}-B_{33})$. It follows from Eq. (12) that a narrow resonance is indicative of strong ME coupling in the composites. Thus NFO-PZT bilayer shows a sharp resonance in comparison to YIG-PZT. Figures 1 and 2 represent the magnetic spectra of the composites obtained by magnetic and electric sweep, respectively, and are of interest for potential use of the composites in devices. Most ferrite based devices use a permanent magnet for the bias field and it is rather difficult to tune them even over a narrow frequency range. It is clear from Fig. 1 that with a ME composite, however, tuning could be accomplished with an easy to generate electric field. For a maximum E value of 300 kV/cm, devices could be tuned over a frequency width varying from a maximum 925 MHz (or $\delta H=330$ Oe) for NFO-PZT to a minimum of 60 MHz ($\delta H=22$ Oe) for YIG-PZT.

III. MAGNETOELECTRIC SUSCEPTIBILITY OF FERRITE-FERROELECTRIC COMPOSITES

Consider now a composite subjected to both dc and ac magnetic and electric fields. According to Eq. (2), it is necessary to consider the relationship between the variable magnetization and the time-dependent electric field for finding the ME susceptibility tensor. Linearized equation of motion for the magnetization for this case can be written as

$$i\omega\mathbf{m}+\gamma\mathbf{m}\times(\mathbf{H}_0-\Sigma N^i\mathbf{M}_0)-\gamma\mathbf{M}_0\times(\mathbf{H}_0-\Sigma N^i\mathbf{m}) \\ =-\gamma\mathbf{M}_0\times\mathbf{h}^e, \tag{13}$$

where $h^e_{n'}=2(B_{ikn}+2b_{ijkn}E_{oj})\beta_{ii}\beta_m\beta_{3k}M_0e_i$.

The magnetic susceptibility in relation to the effective magnetic field $\mathbf{h}^e$ is defined by Eq. (5), i.e.,

$$\mathbf{m}=\chi^m\mathbf{h}^\mathbf{e} \tag{14}$$

From Eq. (5), taking into account Eqs. (13) and (14), it is possible to write the expression for components of ME susceptibility:

$$\chi^{me}_{li}=-2M_0\chi^m_h\eta_{ni}, \tag{15}$$

where $\eta_{ni}=(B_{ikn}+2b_{ijkn}E_{oj})\beta_{ii}\beta_m\beta_{3k}$.

It is evident from the Eq. (15) that the frequency dependence of magnetic and ME susceptibility are identical, and the value of ME susceptibility is determined mainly by ME constants and the dc electric field. Taking into account the internal structure of the magnetic susceptibility tensor, the general expression for ME susceptibility is as follows:

$$\boldsymbol{\chi}^{\mathbf{me}}=\left\| \begin{matrix} \chi^{me}_{11} & \chi^{me}_{12} & \chi^{me}_{13} \\ \chi^{me}_{21} & \chi^{me}_{22} & \chi^{me}_{23} \\ 0 & 0 & 0 \end{matrix} \right\|. \tag{16}$$

It follows from Eq. (16) that, unlike the magnetic susceptibility tensor, $\boldsymbol{\chi}^{\mathbf{me}}$ is generally asymmetric and the third line of the tensor remains zero when magnetic losses are ignored. For practical use of the above expressions we shall consider the case of ME susceptibility tensor for point groups $3m$ and $4mm$. All other point groups with uniaxial structures can be obtained in a similar way. It follows from Eq. (15) that the ME susceptibility tensor can be obtained by multiplication of the magnetic susceptibility tensor in Eq. (5) and tensor **ç**, defined by Eq. (15). Thus for the ME susceptibility tensor components, we get

$$\chi^{ME}_{1'1'} = -D^{-1}\gamma^2 M_0^2(\acute{I}_{03}+2H_a\cos 2\Theta)4[(-B_{22}+b_{66}E_{02}+b_{41}E_{01})\sin\Theta+(B_{15}+b_{14}E_{02}+b_{44}E_{03})\cos\Theta] - i2D^{-1}\gamma M_0^2\omega[(b_{12}-b_{13})E_{01}\sin 2\Theta+2b_{14}E_{01}\cos 2\Theta];$$

$$\chi^{ME}_{1'2'} = -D^{-1}\gamma^2 M_0^2 2(H_{03}+2H_a\cos 2\Theta)[(b_{66}-b_{44})\times E_{01}\sin 2\Theta+2(b_{14}+b_{41})E_{01}\cos^2\Theta-2b_{41}E_{01}] - iD^{-1}\gamma M_0^2\omega 2\{[B_{22}+(b_{11}-b_{13})E_{02}-b_{41}E_{03}]\cos\Theta\times\sin 2\Theta+[B_{33}-B_{31}+b_{41}E_{02}+(b_{33}-b_{31})E_{03}]\sin\Theta\sin 2\Theta + 2(B_{15}+b_{44}E_{03}-b_{14}E_{02})\cos\Theta\cos 2\Theta+2b_{44}E_{02}\sin\Theta\cos 2\Theta\};$$

$$\chi^{ME}_{1'3'} = -D^{-1}\gamma^2 M_0^2 2[H_{03'}+(2H_a-4\pi M_0)\cos 2\Theta][(b_{41}+b_{14})E_{01}\times\sin 2\Theta+2(b_{44}-b_{66})E_{01}\cos^2\Theta+2b_{66}E_{01}] - iD^{-1}\gamma M_0^2\omega 2\{[b_{41}E_{02}-(b_{33}-b_{31})E_{03}]\cos\Theta\times\sin 2\Theta+[(b_{11}-b_{13})E_{02}-b_{41}E_{03}]\sin\Theta\sin 2\Theta + 2b_{44}E_{02}\cos\Theta\cos 2\Theta+q(b_{44}E_{03}-b_{14}E_{02})+2(b_{44}E_{03}-b_{14}E_{02})\sin\Theta\cos 2\Theta\};$$

$$\chi^{ME}_{2'1'} = iD^{-1}\gamma M_0^2\omega 4[(b_{14}E_{03}+B_{66}E_{02})\sin\Theta+(b_{14}E_{02}+B_{44}E_{03})\cos\Theta]-D^{-1}\gamma^2M_0^2[H_{03'}+(2H_a-4\pi M_0)\cos^2\Theta] \times 2[(b_{12}-b_{13})E_{01}\sin 2\Theta+2b_{14}E_{01}\cos 2\Theta];$$

$$\chi^{ME}_{2'2'} = iD^{-1}\gamma M_0^2\omega r[(b_{66}-b_{44})E_{01}\sin 2\Theta+2(b_{14}+b_{41})E_{01}\cos^2\Theta-4b_{41}E_{01}]-D^{-1}\gamma^2M_0^2[H_{03'}+(2H_a-4\pi M_0)\cos^2\Theta] \times\{[2(b_{11}-b_{13})E_{02}-2b_{41}E_{03}]\cos\Theta\times\sin 2\Theta+2[b_{41}E_{02}+(b_{33}-b_{31})E_{03}]\sin\Theta \times\sin 2\Theta+4(-b_{14}E_{02}+b_{44}E_{03})\cos\Theta\cos 2\Theta-4b_{44}E_{02}\sin\Theta\cos 2\Theta\}; \tag{17}$$

$$\chi^{ME}_{2'3'} = -D^{-1}\gamma M_0^2\omega[2(b_{14}+b_{41})E_{01}\sin 2\Theta+4(b_{44}-b_{66})E_{01}\cos^2\Theta+4b_{66}E_{01}]-D^{-1}\gamma^2M_0^2[H_{03'}+(2H_a-4\pi M_0)\cos^2\Theta] \times\{[-b_{41}E_{02}-(b_{33}-b_{31})E_{03}]\cos\Theta\sin 2\Theta+2[(b_{11}-b_{13})E_{02}-b_{41}E_{03}]\sin\Theta\sin 2\Theta+4b_{44}E_{02}\times\cos\Theta\cos 2\Theta + 4(b_{44}E_{03}-b_{14}E_{02})\sin\Theta\cos 2\Theta\},$$

where

$$D=\omega_0^2-\omega^2;$$

$$\omega_0^2=\gamma^2[H_{03'}+2(H_a-2\pi M_0+q_1M_0)\cos^2\Theta+2M_0\sin 2\Theta + M_0q_2]\times[H_{03'}+2(H_a-2\pi M_0+q_1M_0)\cos 2\Theta + 4rM_0\sin 2\Theta];$$

$$q_1=(b_{12}-b_{13})E_{01}^2+(b_{11}-b_{13})E_{02}^2+(b_{31}-b_{33})E_{03}^2 - 2b_{41}E_{02}E_{03};$$

$$r=b_{14}(E_{02}^2-E_{01}^2)-2b_{44}E_{02}E_{03};$$

$$q_2=r(b_{11}-b_{12})(E_{02}^2-E_{01}^2)+8b_{41}E_{02}E_{03}.$$

Now we consider some special cases having practical importance and simplify these expressions. Without loss of generality, we shall consider a case when the equilibrium magnetization coincides with a symmetry axis of a structure. In that case in the Eq. (17) we assume $\Theta=0$ and $\acute{I}_0=\acute{I}_{03}$. For the ME susceptibility tensor components, we obtain

$$\chi^{ME}_{1'1'} = -2D^{-1}\gamma^2M_0^2(H_0+2H_a)[B_{15}+2(b_{25}E_{01}+\lambda b_{14}E_{02} + b_{44}E_{03})]-2iD^{-1}\gamma M_0^2\omega[B_{14}+2(b_{14}E_{01}+b_{25}E_{02} - b_{45}E_{03})];$$

$$\chi^{ME}_{1'2'} = -2D^{-1}\gamma^2M_0^2(H_0+2H_a)[-B_{14}+2(b_{14}-b_{41})E_{01} + 2(b_{25}-b_{53})E_{02}+2b_{45}E_{03}]-2iD^{-1}\gamma M_0^2\omega[B_{15} + 2(b_{25}E_{01}-b_{14}E_{02}+b_{44}E_{03})];$$

$$\chi^{ME}_{1'3'} = -D^{-1}\gamma^2M_0^2(H_0+2H_a)[2(b_{44}E_{01}+b_{45}E_{02})] - 4iD^{-1}\gamma M_0^2\omega(B_{44}E_{02}-b_{45}E_{01});$$

$$\chi^{ME}_{2'1'} = +2iD^{-1}\gamma^2 M_0^2\omega[B_{15}+2(b_{25}E_{01}+b_{14}E_{02}+b_{44}E_{03})] - 2D^{-1}\gamma^2 M_0^2(H_0+2H_a)[B_{14} + 2(b_{14}E_{01}+b_{25}E_{02}-b_{45}E_{03})];$$

$$\chi^{ME}_{2'2'} = -2iD^{-1}\gamma M_0^2\omega[-B_{14}+2(b_{14}-b_{41})E_{01}-2(b_{25} - b_{52})E_{02}+2b_{45}E_{03}] - 2D^{-1}\gamma^2 M_0^2[B_{15}+2(b_{25}E_{01} - b_{14}E_{02}+b_{44}E_{03})](H_0+2H_a); \quad (18)$$

$$\chi^{ME}_{2'3'} = +4iD^{-1}\gamma M_0^2\omega[b_{44}E_{01}+b_{45}E_{02}] - 4D^{-1}\gamma^2 M_0^2 \times(H_0+2H_a)(b_{44}E_{02}-b_{45}E_{01}),$$

where

$$D = \gamma^2[H_0+2H_a+2q_1M_0+M_0q_2][H_0+2H_a+2q_1M_0] - \omega^2;$$

q_1 and q_2 are determined by Eq. (17). We shall consider a disk sample cut in a base plane with electrical and magnetic-field directions perpendicularly to the disk plane ($\mathbf{M_0}\|\mathbf{E_0}\|\mathbf{3}$). Then from Eq. (18), we get

$$\chi^{ME}_{1'1'} = \chi^{ME}_{2'2'} = -2D^{-1}\gamma^2 M_0^2(H_0+2H_a-4\pi M_0) \times(B_{15}+2b_{44}E_0),$$

$$\chi^{ME}_{1'2'} = -\chi^{ME}_{2'1'} = -2iD^{-1}\gamma M_0^2\omega(B_{15}+2b_{44}E_0), \quad (19)$$

$$\chi^{ME}_{1'3'} = \chi^{ME}_{3'1'} = 0,$$

where

$$D = \gamma^2[H_0+2(H_a+M_0q-2\pi M_0)]^2 - \omega^2;$$

$$q = (B_{31}-B_{33})E_0 + (b_{31}-b_{33})E_0^2.$$

From Eq. (19) it is evident that in this case the ME susceptibility tensor is antisymmetric and can be diagonalized. For this purpose we use cyclic variables $e_\pm = e_{1'} \pm ie_{2'}$, $m_\pm = m_{1'} \pm im_{2'}$. So, we obtain

$$\chi^{ME}_\pm = -2\chi^M_\pm M_0(B_{15}+2b_{44}E_0), \quad (20)$$

where $\chi^M_\pm = \chi_1 \pm \chi_a$.

Equation (20) facilitates direct estimation of the ratio of magnetoelectric and magnetic susceptibilities. We consider here a specific example: bulk composites of YIG and PZT. Using the experimental data on linear and bilinear ME effect and taking into account that $B_{15} \approx B_{33} - B_{13}$, we get $2B_{15}M_0 = 0.3$ Oe cm/kV $= 9\times10^{-2}$ in Gaussian units.[24] Assuming negligible bilinear terms and substituting the value of B_{15} in Eq. (20), we get

$$\left|\frac{\chi^{ME}_\pm}{\chi^M_\pm}\right| = 0.3\,\frac{\text{Oe cm}}{\text{kV}}.$$

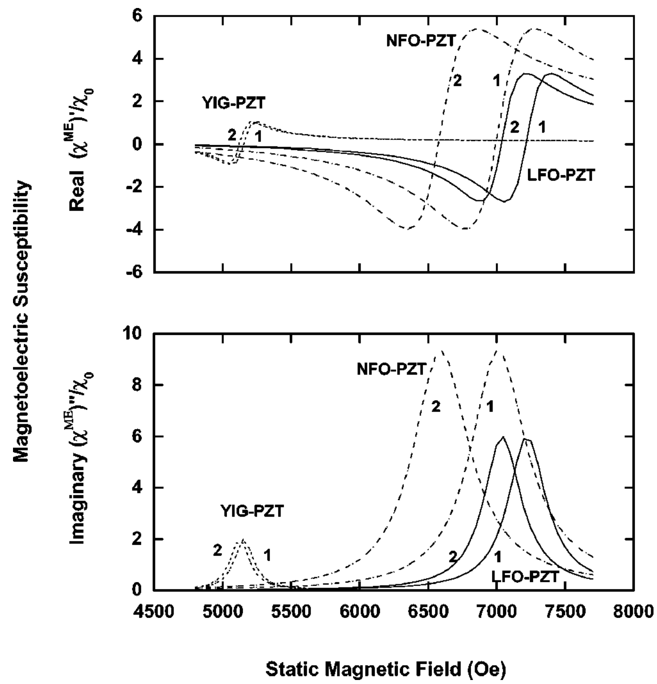

FIG. 3. Theoretical magnetic-field dependence of the magnetoelectric (ME) susceptibility for bilayer composites of LFO-PZT, NFO-PZT, and YIG-PZT at 9.3 GHz. Both real and imaginary components are shown for (1) $E=0$ and (2) $E=300$ kV/cm.

If the linear term is negligible, as is the case for unpolarized YIG-PZT samples, and with the bilinear ME constant $4b_{44}M_0 = 0.7\times10^{-2}$ Oe (cm/kV)$^2 = 6.3\times10^{-4}$ esu^{-1} and for $Å_0 = 1$ kV/cm, we find

$$\left|\frac{\chi^{ME}_\pm}{\chi^M_\pm}\right| = 0.7\times10^{-2}\frac{\text{Oe cm}}{\text{kV}}.$$

Magnetic- or electric-field profiles of ME susceptibility could be obtained from Eqs. (12a) and (15). As an example, we once again consider bilayers of LFO-PZT, NFO-PZT, and YIG-PZT. Figure 3 shows the calculated dependence of real and imaginary parts of ME susceptibility on static magnetic field. The results are for a frequency of 9.3 GHz, as for Fig. 1, and $E=0$ and 300 kV/cm. Important features in Fig. 3 are essentially similar to the electric-field effect on magnetic susceptibilities discussed in Sec. II. A resonance is seen in the susceptibility variation with H. A downshift in H_r accompanies the application of E. Since the amplitude for ME susceptibility is determined by the ME constants, we observe in Fig. 3 a larger amplitude for NFO-PZT than for YIG-PZT. It is worth noting that Fig. 3 represents a unique property for ME composite and can be defined as ME spectrum of composite.

IV. DISCUSSION

Here we describe briefly experimental techniques for the determination of magnetic and ME susceptibilities. We then employ an indirect procedure, measurements of electric-field induced ferromagnetic resonance (FMR) line shift for the estimation of ME constants for multilayers of LFO-PZT. The parameters thus obtained are then used to calculate the susceptibilities. The microwave cavity resonance technique is ideal for the determination magnetic susceptibility. In this procedure, the sample is mounted at the maximum of rf magnetic field. One needs to measure the shift in the resonance frequency and the cavity Q, quantities that are proportional to the real and imaginary parts of the magnetic susceptibility. Measurements as a function of H and E are necessary for field profiles for the susceptibility as in Figs. 1 and 2.

There are three methods for measurements of microwave ME susceptibilities in ferrite-PZT composites:[21] (i) resonant coupling of microwave magnetic and electrical fields due to ME interaction in the material; (ii) electric dipole transitions in the composite; (iii) ferromagnetic resonance line shift due to a dc electric field and estimation of ME constants and susceptibilities. The first two are direct procedures and the third is an indirect but effective method. In the first method, input and output antenna made of transmission lines or resonators are coupled through a sample of the composite of interest. The sample is required for the transformation of microwave magnetic field into electric field or vice versa. For the determination of ME susceptibility by power absorption at electric dipole transitions, the sample is located at the maximum of microwave electric field in a microwave resonator. The absorbed power is proportional to an effective microwave magnetic field resulting from ME interactions and the sample geometry. The determination of ME susceptibilities requires the amplitude of rf electric field and the power absorbed.[21]

The third measurement technique is an indirect procedure. The value of ME susceptibility is determined by ME constants. For theoretical susceptibilities in Figs. 1–3, we calculated ME constants from material parameters for the ferrites and PZT. But one could also determine the coupling constants experimentally from data on magnetic resonance field shift δH due to constant electric fields. We employ this procedure here for a specific case, multilayer composites of LFO-PZT. Multilayer composites of LFO-PZT were prepared from 15-μm-thick films of the ferrite and PZT made by tape casting techniques.[26] Samples containing 16 layers of LFO and 15 layers of PZT were prepared by lamination and sintering at 1450 K. Further details on sample preparations and low-frequency characterization will be discussed elsewhere.[27] The composites were polished and electrical contacts were made with silver paint. Ferromagnetic resonance studies using a microstripline structure operating at 9.3 GHz were carried out at room temperature on discs of diameter 4 mm and thickness 0.5 mm. The static magnetic field was applied perpendicular to the sample plane and power absorption by the sample was measured as a function of H. Resonance was observed with a linewidth was on the order of 300 Oe. The samples were subjected to a constant electric field perpendicular to its plane and the resonance absorption versus static magnetic-field profile was obtained for a series of electric fields. Figure 4 shows the variation of the shift δH in the resonance field as a function of E. A linear dependence of the field shift on the electric field is evident from the data and is indicative of the absence of measurable bilinear ME effects. One obtains, from data in Fig. 4, a linear ME coefficient such that $2M_o(B_{31}-B_{33})=0.4$ Oe cm/kV. From Eq. (20) we get the ratio of magnetoelectric and magnetic susceptibilities for LFO-PZT multilayer composite,

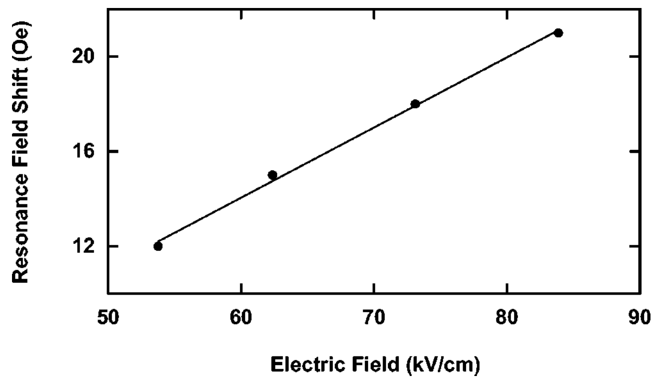

FIG. 4. Resonant magnetoelectric effect measured in a multilayer composite of LFO-PZT. The sample contained 16 layers of LFO and 15 layers of PZT. The thickness of each layer is 15 μm. The static fields H and E are perpendicular to the sample plane. The shift in the ferromagnetic resonance field at 9.3 GHz is shown as a function of E. The ME constants obtained from the data are used to calculate the magnetic and ME susceptibilities for the multilayer.

$$\left|\frac{\chi_{\pm}^{ME}}{\chi_{\pm}^{M}}\right|=0.4\,\frac{\text{Oe cm}}{\text{kV}}.$$

The ratio is in very good agreement with the theoretical estimate of 0.6 Oe cm/kV obtained in Sec. III for a bilayer of LFO-PZT. Similar data, but for other combinations of E and H field orientations, could be used for the determination of other ME constants of interest.

The ME constants thus obtained is useful for the calculation of susceptibilities for the multilayer composite. Figure 5 shows the variation of the real and imaginary parts of the magnetic susceptibility for the LFO-PZT sample as a function of H and E for a frequency of 9.3 GHz. Other parameters used in the calculation are $\alpha=0.03$ obtained from FMR data and $H_0+2H_a-4\pi M_0=3300$ Oe. These estimations are made for a disk sample magnetized along the symmetry axis. Results on χ vs H in Fig. 5 reveal an E-induced shift δH of 117 Oe, in agreement with 136 Oe for the bilayer of LFO-PZT. Similarly, the width of χ vs E profile in Fig. 5 agrees well with the estimates for the bilayer in Fig. 2. The ME susceptibility and its dependence on H and E for the multilayer sample is shown in Fig. 6. Comparison with results for the bilayer in Fig. 3 indicates small amplitude of resonance for the multilayer and is due to weak magnetoelectric interactions. Thus results in Figs. 5 and 6 provide the essential information on high-frequency magnetic and ME susceptibilities for LFO-PZT multilayers. The electric-field influence on the magnetic susceptibilities in LFO-PZT is stronger than in YIG-PZT, but weaker compared to NFO-PZT.

As mentioned in the introduction, ME devices based on the interaction between rf magnetic field and constant E field [type (i)] are of interest in the microwave region of the elec-

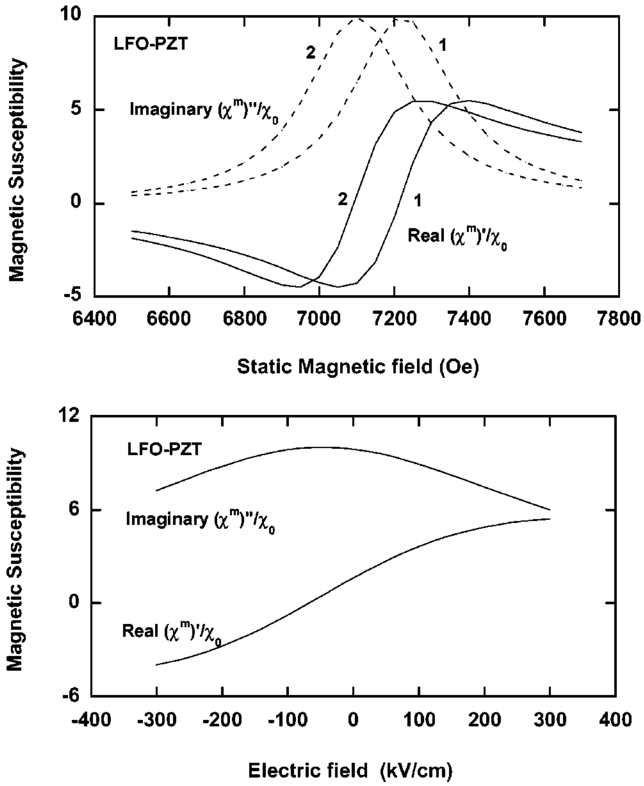

FIG. 5. Magnetic-field and electric-field dependence of the magnetic susceptibility for the multilayer of LFO-PZT. The susceptibilities at 9.3 GHz and (1) $E=0$ and (2) $E=300$ kV/cm were estimated using ME constants obtained from the resonance field shift data in Fig. 4.

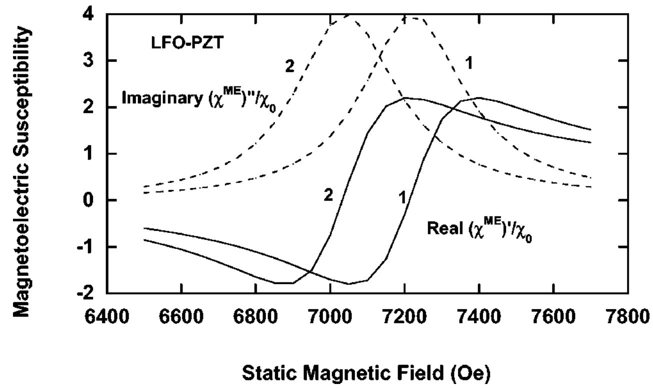

FIG. 6. Magnetic-field dependence of the ME susceptibility for the multilayer composite of LFO-PZT. The estimated values at 9.3 GHz are for ME constants obtained from data in Fig. 4 and for (1) $E=0$ and (2) $E=300$ kV/cm.

tromagnetic spectrum. The E induced shift in the resonance field in such materials should at least equal the resonance linewidth. It is clear from Figs. 1, 2, and 5 that all the composites considered here satisfy the key requirement. Results in Fig. 1 indicate the suitability of NFO-PZT composites for type-I devices such as phase shifters, and attenuators, as the line shift is 330 Oe in $E=300$ kV/cm versus a resonance linewidth of 450 Oe. Results of ME susceptibilities in Figs. 3 and 6 are indicative of potential use of the composites for type (ii) devices such as coupled wave devices, parametric oscillators, and transformers.

V. CONCLUSIONS

General expressions for the electric-field effects on magnetic and ME susceptibility tensors of ferroelectric/magnetostrictive composite (symmetry point group 3 m and 4 mm) at microwave frequencies have been obtained. Expressions for the magnetic susceptibility indicate that the static electric field does not alter the tensor structure and one expects a resonance in the electric-field dependence. Similarly, the ME susceptibility tensor has resonant dependence on external magnetic and electric fields. The value of ME susceptibility is determined by ME constants and the electric field. The theory is then used to calculate the susceptibilities for bilayer composites of LFO-PZT, NFO-PZT and YIG-PZT. One observes important characteristics such as strong E-field influence and a sharp resonance in the susceptibility vs E profile for NFO-PZT. We proposed two direct measurement techniques for ME susceptibility, resonant ME coupling, and electrical dipole transitions. An indirect procedure based on ME effects at ferromagnetic was used to obtain ME constants for subsequent calculation of the susceptibilities for multilayer samples of LFO-PZT.

The theory presented here is of fundamental and technological importance. It enables the determination of the strength of ME interactions from appropriate data and subsequent prediction of susceptibilities for composites such as NFO-PZT, LFO-PZT, and YIG-PZT. The theory is also of importance for the design and the analysis of devices based on ME composites.

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