Effect of inclusion deformation on the magnetoelectric effect of particulate magnetostrictive/piezoelectric composites

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We have investigated the magnetoelectric (ME) properties of particulate composites with magnetostrictive inclusions dispersed in a piezoelectric matrix. A simple model is proposed which includes a rarely discussed contribution from the shape deformation of the inclusion particles due to the magnetostrictive stresses generated under the action of an external magnetic field. The relative significance of this contribution is mainly determined by the ratio of the transverse and longitudinal magnetostrictive responses of the inclusion as well as the flexibility of the piezoelectric matrix phase. This model gives further insight on material selections for designing particulate ME composites. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2781513]

I. INTRODUCTION

Magnetoelectric (ME) composites formed a subject of interest in recent years because coupled magnetostrictive-piezoelectric effect in a two-phase composite can exhibit a much higher ME effect than that of a single-phase intrinsic ME material. The ME effect in an ME composite occurs as a result of the product property between the magnetostrictive and piezoelectric responses.[1] In the literature, both particulate and laminated composites have been developed. Many theoretical investigations of the ME effect are focused on laminated structures,[1–5] which requires a less cumbersome treatment than particulate composites. Some sophisticated models for particulate composites do exist,[6–8] but their approaches are rather complicated and heavy numerical computation schemes are normally employed for calculating the ME coefficients. Also, the nonlinear constitutive relationships for magnetostriction and magnetization-magnetic field in the magnetostrictive phase place difficulties for developing simple models for the ME composites. In attempts to model such magnetostriction and magnetization behaviors with simple analytical expressions, the works of Carman *et al.*[9–11] and Zheng *et al.*[11,12] may be cited, in which their results are convenient to be used.

Previously, we have proposed simple models for the effective piezoelectric properties of 0–3 composites and the longitudinal ME response for dilute particulate magnetostrictive-piezoelectric composites with spherical inclusions.[13,14] Fairly good agreement with experimental results has been obtained. To impart piezoelectric activity in a 0–3 ferroelectric composite, the material must first be subjected to a poling process for a sufficiently long duration to align the spontaneous polarization as well as to allow free charges to accumulate at the inclusion-matrix interfaces to counteract the depolarization field to stabilize the polarization. In our studies of piezoelectric composites, we note that the effect of inclusion deformation, due to the applied stress in piezoelectric measurement, can have strong implications on the gross piezoelectric properties of composites with oppositely polarized constituents when the presence of these compensating free charges at the interfaces is taken into account.[15] Likewise, deformation of the magnetostrictive particles also occurs in an ME composite, caused by the magnetostrictive stresses due to the applied magnetic field. Since the ferroelectric matrix phase must also be prepolarized to acquire piezoelectric activity to give an ME response, the existence of interfacial charge in the particulate composite sample is thus an essential fact that should not be overlooked in this context as it may also have significant implications on the ME behavior. The concomitant inclusion deformation effect on the ME properties has not been examined before.

In this work, we attempt to study theoretically the ME responses of dilute particulate composites comprising magnetostrictive inclusions dispersed in piezoelectric matrix. The deformation of the inclusion volume and hence the charged interface due to magnetostrictive stress is taken into consideration. Following our previous assumptions that the constituent materials are dielectrically and elastically isotropic, both longitudinal (α_{33}) and transverse (α_{31}) ME responses are studied. Our theoretical predictions will first be compared with the published experimental results of α_{33} for $NiFe_2O_4$/lead zirconate titanate (NFO/PZT) composites.[16] Then the ME responses of the particulate composites with vinylidene fluoride-trifluoroethylene [P(VDF-TrFE)] copolymer as piezoelectric matrix will be investigated. The latter is considered to be a good candidate for the piezoelectric phase of ME composites,[6,17,18] since P(VDF-TrFE) possesses large values of piezoelectric *g* coefficients and has the advantage of high voltage output due to its low permittivity value.[18]

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II. THEORY

In the configuration of the biphasic particulate ME composite such as prepared by Zhai *et al.*,[16] the magnetostrictive inclusions are embedded in a piezoelectric matrix. We will first give a general formulation for the ME coefficient α of the composite in dilute limit under an external magnetic field, then calculations for transverse and longitudinal magnetic fields will be discussed.

A. Magnetoelectricity of a composite with inclusion deformation effect

We first write the volumetric average electric displacement for the ferroelectric constituent materials in the composite as $D_j=\varepsilon_j\ E_j+P_j$ where symbol D denotes electric displacement, ε is permittivity, E is electric field, and P is polarization. The subscript $j=i$ for inclusion and $j=m$ for matrix. In this article, we are interested in the composite of nonferroelectric inclusions in a ferroelectric matrix only, thus $P_i=0$.

Consider the single inclusion problem of a dielectric sphere surrounded by a ferroelectric matrix medium with a uniform electric field applied along the z direction far away from the inclusion. The boundary value problem gives the following equation:[15]

$$D_i+2\varepsilon_m(E_i-E_m)=D_m-q_0. \tag{1}$$

In Eq. (1), we have assumed the matrix material is uniformly polarized and the homogeneously polarized sphere is covered with surface charge of density q_0 at the pole along the polarizing direction ($\theta=0$) with a distribution given by $q_0\cos\theta$.

For a composite comprising a dilute suspension of spherical particles uniformly distributed in the matrix material, the volumetric averages of the electric field and electric displacement are[13,15]

$$\begin{aligned} E&=\phi E_i+(1-\phi)E_m,\\ D&=\phi D_i+(1-\phi)D_m. \end{aligned} \tag{2}$$

ϕ is the volume fraction of the inclusion phase. Using Eqs. (1) and (2), we obtain, after some algebraic manipulation

$$D=\varepsilon E+(1-\phi)\bar{L}_E P_m+\phi(1-\phi)(L_E-\bar{L}_E)q_0, \tag{3}$$

where $\varepsilon=\varepsilon_m\{(\varepsilon_i+2\varepsilon_m)+2\phi(\varepsilon_i-\varepsilon_m)\}/\{(\varepsilon_i+2\varepsilon_m)-\phi(\varepsilon_i-\varepsilon_m)\}$ and

$$\begin{aligned} L_E&=\frac{3\varepsilon_m}{(1-\phi)\varepsilon_i+(2+\phi)\varepsilon_m},\\ \bar{L}_E&=\frac{1-\phi L_E}{1-\phi}=\frac{\varepsilon_i+2\varepsilon_m}{(1-\phi)\varepsilon_i+(2+\phi)\varepsilon_m}. \end{aligned} \tag{4}$$

When stresses are also present in the system, then assuming permittivities ε_i and ε_m, and the charge density q_0 do not vary with stress, the change of D can be evaluated by[15]

$$\Delta D=\frac{\partial D}{\partial E}\Delta E_i+\frac{\partial D}{\partial P_m}\Delta P_m+\frac{\partial D}{\partial \phi}\Delta\phi, \tag{5}$$

where

$$\Delta P_m=d_{31m}T_{xm}+d_{32m}T_{ym}+d_{33m}T_{zm}, \tag{6}$$

$$\Delta\phi=\phi(\gamma_i-\gamma). \tag{7}$$

d_{31}, d_{32}, and d_{33} are piezoelectric coefficients. γ is the volumetric strain which is the summation of tensile strain components, i.e., $\gamma=e_x+e_y+e_z$ for the composite and similarly γ_i for the inclusions. T_x and e_x represent the tensile stress and strain in the x direction, respectively. To facilitate the subsequent calculations, Eq. (7) may be transformed to $\Delta\phi=\phi(1-\phi)(\gamma_i-\gamma_m)$ using the relation $\gamma=\phi\ \gamma_i+(1-\phi)\gamma_m$.

The ME coefficient α_{3f} is defined as $t^{-1}dV/dH$, where V, H, and t denote output voltage, applied magnetic field, and the sample thickness, respectively. The first index "3" addresses the poling direction (assumed along z direction) of the composite and the second index "f" (=1, 2, or 3) denotes the direction of magnetic field. At short circuit condition, $E=\Delta E=0$, and the output voltage is obtained via the measurement of charge generated from the sample (denoted by Q) with $V=Q/C$, where C is the capacitance of the sample. Since $\Delta D=Q/A$ and $C=\varepsilon A/t$, we differentiate Eq. (5) once with H to get

$$\begin{aligned} \alpha_{3f}&=(1-\phi)\frac{\bar{L}_E}{\varepsilon}\left(d_{31m}\frac{dT_{xm}}{dH_i}+d_{32m}\frac{dT_{ym}}{dH_i}+d_{33m}\frac{dT_{zm}}{dH_i}\right)\\ &\quad\times\left(\frac{dH_i}{dH}\right)+\alpha_\phi, \end{aligned} \tag{8}$$

where

$$\begin{aligned} \alpha_\phi&=\varepsilon^{-1}\frac{d(\Delta\phi)}{dH_i}\left\{-\left[\bar{L}_E-(1-\phi)\frac{\partial\bar{L}_E}{\partial\phi}\right]P_{rm}\right.\\ &\quad\left.+\left[(1-2\phi)(L_E-\bar{L}_E)+\phi(1-\phi)\frac{\partial(L_E-\bar{L}_E)}{\partial\phi}\right]q_0\right\}\\ &\quad\times\left(\frac{dH_i}{dH}\right), \end{aligned} \tag{9}$$

and

$$\frac{\partial\bar{L}_E}{\partial\phi}=\frac{(\varepsilon_i+2\varepsilon_m)(\varepsilon_i-\varepsilon_m)}{[(1-\phi)\varepsilon_i+(2+\phi)\varepsilon_m]^2}. \tag{10}$$

We assumed all internal electric fields vanish initially. Hence we have set $P_m=P_{rm}$ (i.e., remanent polarization of the matrix phase) in Eq. (9). We also have $q_0=P_{rm}$ since $q_0=P_m-P_i+3\varepsilon_m E_m-(\varepsilon_i-\varepsilon_m)E_i$ [obtained from Eq. (1)] and $P_i=E_i=E_m=0$. In other words, the polarization in the matrix phase is initially fully compensated by the free-surface charge. α_ϕ [Eq. (9)] is the contribution arising from the change in inclusion volume fraction with the embedded interfacial charge effect. Thus the consideration of change in ϕ due to stress also leads to an interfacial charge effect in the ME response.

The derivative of H_i with respect to H [in Eqs. (8) and (9)] can be obtained by first considering the constitutive magnetostatic equations

$$B_i = \xi_i H_i + M_i, \qquad B_m = \xi_m H_m, \tag{11}$$

where B is the magnetic flux density, ξ is the magnetic permeability, and M is the magnetization. Normally we can take $\xi_m = \xi_0$ for nonmagnetic piezoelectric matrix. Since the particulate composite is subjected to an applied magnetic field, we have

$$H = \phi H_i + (1-\phi)H_m. \tag{12}$$

The boundary conditions of the magnetic field and magnetic flux density gives the following equation:[19]

$$B_i + 2\xi_m(H_i - H_m) = B_m. \tag{13}$$

Combining Eqs. (11)–(13) we obtain

$$\frac{dH_i}{dH} = \frac{2\xi_m}{(1-\phi)(\xi_i + dM_i/dH_i) + (2+\phi)\xi_m}. \tag{14}$$

Thus for a given external field H acting on the composite, we may obtain dH_i/dH when the M_i-H_i relation is known. The model suggested by Duenas, Hsu, and Carman may be adopted in this calculation[10,11]

$$M_i = M_{\mathrm{sat},i}\tanh(3\chi_i H_i/M_{\mathrm{sat},i}), \tag{15}$$

where M_{sat} is the saturation magnetization and χ is a relaxation factor.

B. Internal stresses and strains in the particulate composites

Equation (8) requires the values of $\gamma_i(=e_{xi}+e_{yi}+e_{zi})$, γ_m, T_{xm}, T_{ym}, and T_{zm}. They can be evaluated by solving appropriate elasticity equations and boundary conditions. Suppose both phases of the composite are elastically isotropic, the stress and strain in the x, y, and z directions of the constituents obey the following relationship:

$$\begin{aligned} e_{xi} &= T_{xi}/Y_i - \nu_i(T_{yi}+T_{zi})/Y_i + \lambda_{xi}(H_i),\\ e_{yi} &= T_{yi}/Y_i - \nu_i(T_{xi}+T_{zi})/Y_i + \lambda_{yi}(H_i),\\ e_{zi} &= T_{zi}/Y_i - \nu_i(T_{xi}+T_{yi})/Y_i + \lambda_{zi}(H_i), \end{aligned} \tag{16}$$

$$\begin{aligned} e_{xm} &= T_{xm}/Y_m - \nu_m(T_{ym}+T_{zm})/Y_m,\\ e_{ym} &= T_{ym}/Y_m - \nu_m(T_{xm}+T_{zm})/Y_m,\\ e_{zm} &= T_{zm}/Y_m - \nu_m(T_{xm}+T_{ym})/Y_m. \end{aligned} \tag{17}$$

In Eqs. (16) and (17), Y and ν denote Young's modulus and Poisson's ratio, respectively. λ_{xi}, λ_{yi}, and λ_{zi} are the magnetic field induced strains in the inclusions due to H_i. For a homogeneous magnetostrictive inclusion, the variations of λ_i's with H_i may either be taken from experimental data or be conveniently evaluated by models for magnetostrictive response, while the instantaneous H_i is obtained by solving Eqs. (11)–(13).

In a previous article, we have considered the elastic boundary value problem which can give the relationships between the inclusion and matrix stresses and strains.[13] The results can be rewritten as

$$\begin{pmatrix} T_{xi}-T_{xm}\\ T_{yi}-T_{ym}\\ T_{zi}-T_{zm}\end{pmatrix} = \begin{pmatrix} A & B & B\\ B & A & B\\ B & B & A\end{pmatrix}\begin{pmatrix} e_{xi}-e_{xm}\\ e_{yi}-e_{ym}\\ e_{zi}-e_{zm}\end{pmatrix}, \tag{18}$$

where $A=10\mu_m\{2\mu_m/(k_m+2\mu_m)-3\}/9$ and $B=-\mu_m\{10\mu_m/(k_m+2\mu_m)+3\}/9$.[19] Symbols k and μ denote bulk modulus and shear modulus, respectively. In this work, we focus on the case that the composite is not clamped in either x, y, or z directions. Since the overall transverse and longitudinal stresses should be balanced

$$T_\ell = \phi T_{i\ell} + (1-\phi)T_{m\ell} = 0, \tag{19}$$

where ℓ can be x, y, or z. Using Eqs. (16)–(19), the expressions for γ_i, γ_m, T_{xm}, T_{ym}, and T_{zm} can be obtained. Substitution of the resulting expressions for γ_i and γ_m into Eq. (7) gives

$$\Delta\phi = \phi(1-\phi)\frac{3(\lambda_{xi}+\lambda_{yi}+\lambda_{zi})k_i k_m}{3k_i k_m + 4[\phi k_i + (1-\phi)k_m]\mu_m}. \tag{20}$$

C. Magnetoelectric α_{33} and α_{31} coefficients of the composites

When H is applied in the z direction, $\alpha_{3f}=\alpha_{33}$ and we may write $\lambda_i^{\parallel}\equiv\lambda_{zi}$, and $\lambda_i^{\perp}\equiv\lambda_{xi}=\lambda_{yi}$ in Eq. (16) to represent the magnetostriction in the inclusions parallel and perpendicular to H_i, respectively. Hence, $e_{xi}=e_{yi}$, $e_{xm}=e_{ym}$, $T_{xi}=T_{yi}$, and $T_{xm}=T_{ym}$ in Eqs. (8) and (9).

Concerning the ME effect with "transverse" H in which the applied magnetic field is in a transverse direction (say, x direction) with the electric signal measured in the z direction, then the ME coefficient α_{31} is involved. Therefore, $\alpha_{3f}=\alpha_{31}$ in Eq. (8) and $\lambda_{xi}=\lambda_i^{\parallel}$ and $\lambda_{yi}=\lambda_{zi}=\lambda_i^{\perp}$.

Following Duenas, Hsu, and Carman,[10,11] we adopted the expression

$$\lambda_i = \lambda_{\mathrm{sat},i}\tanh^2(3\chi_i H_i/M_{\mathrm{sat},i}) \tag{21}$$

to describe $\lambda_i^{\parallel}$, where $\lambda_{\mathrm{sat},i}$ is the saturated value of magnetostriction. For $\lambda_i^{\perp}$ in our subsequent calculations, we assume $\lambda_i^{\perp}=-\beta\,\lambda_i^{\parallel}$, where β is a positive constant which we call the magnetostrictive strain ratio, for simplicity.

III. RESULTS AND DISCUSSION

A. Comparison with experimental data

In this section, theoretical predictions based on the foregoing model are investigated. We will first concentrate on the calculation of the NFO/PZT system of Zhai *et al.*[16] They have measured the ME coefficient α_{33}. The permeability, permittivity, bulk and shear moduli, remanent polarization, and the piezoelectric coefficients of the constituents adopted for the calculations are shown in Table I. Concerning the parameters in the M_i-H_i [Eq. (15)] and $\lambda_i^{\parallel}$-H_i [Eq. (21)] pro-

TABLE I. Properties of constituents for the NFO/PZT and NFO/P(VDF-TrFE) particulate composites.

	ξ	$\varepsilon/\varepsilon_0$	k (GPa)	μ (GPa)	P_r (μC/cm^2)	d_{33} (pC/N)	$-d_{31}$ (pC/N)
NFO	3[a]	10[b]	197[a]	56[a]	0	0	0
PZT	1	1560[c]	62.3[a]	27.1[a]	33[d]	375[c]	175[a]
P(VDF-TrFE)	1	9.5[d]	2.16[d]	0.5[d]	5.6[d]	−37 [d]	−16 [d]

[a]Reference 14.
[b]Reference 20.
[c]Reference 16.
[d]Reference 15.

files, the article of Zhai *et al.* has not given the magnetic and magnetostriction behavior of NFO. We have set $M_{\mathrm{sat},i}$ $=135$ emu/cm^3,[21] $\lambda_{\mathrm{sat},i}=-30$ ppm,[22] and $\chi_i=0.07$ emu Oe^{-1} cm^{-3}, in the model of Duenas, Hsu, and Carman [Eqs. (15) and (21)]. We first assume the magnetostrictive strain ratio $\beta=1/2(=-\lambda_i^{\perp}/\lambda_i^{\parallel})$ which eventually leads to $\Delta\phi=0$ [since $\lambda_{xi}=\lambda_{yi}=\lambda_i^{\perp}=-\lambda_i^{\parallel}/2$ and $\lambda_{zi}=\lambda_i^{\parallel}$, see Eq. (20)] and $\alpha_\phi=0$ [see Eq. (9)]. In other words, the effect of inclusion deformation vanishes when $\beta=1/2$. Figure 1(a) shows the comparison of theoretical predictions with the α_{33} values of the NFO/PZT composites. In Fig. 1(a), solid, dashed, and dotted lines are based on the theoretical predictions of ϕ $=0.07$, 0.2, and 0.32, respectively. The scatter points are the experimental data with different symbols representing different volume fractions. It is found that fairly good agreement is obtained. However, one may note that the fit is good at medium H region, but not so good at lower and higher H regions. Zhai *et al.*'s experimental data revealed that the peak value of the α_{33}-H curve glides slightly toward a higher H for increasing ϕ. Also, their measurement on the M-H and ME behaviors of the NFO/PZT composites revealed some hysteretic nature (with the loop size increasing with ϕ in M-H), suggesting that the gliding phenomenon may be due to the hysteretic nature in the magnetic and magnetostriction behaviors of NFO. The enhancement of hysteresis with ϕ may very well come from a variation of M with stress which, in the case of a magnetostrictive inclusion particle deformed by a magnetic field, arises from contact with the matrix material. This characteristic is not included in the adopted M_i-H_i [Eq. (15)] and $\lambda_i^{\parallel}$-H_i [Eq. (21)] models of Duenas, Hsu, and Carman. A more sophisticated model may be employed for the $\lambda_i^{\parallel}$-H_i and $\lambda_i^{\perp}$-H_i relationships to give an improved fitting, otherwise numerical approximation for $\lambda_i^{\parallel}$-H_i and $\lambda_i^{\perp}$-H_i can be adopted if the experimental λ_i-H_i curves are measured. However, our focus here is on the effect of inclusion deformation in the ME effect of particulate composites, a simple analytical model for λ_i-H_i will suffice for illustration, else the application of our model would become unnecessarily complicated. Since the present model is primarily for composites with dilute suspension, the predictions at higher $\phi(=0.32)$ seem to be an overestimation for the field region of peak α values [see Fig. 1(a)]. Another possible cause is the effect of electrical conductivities in the constituents, which allow charge accumulation at the inclusion-matrix interfaces, thus affecting the electric field in the piezoelectric matrix.[14] The previous article has suggested that the effective conductivity may increase rapidly with ϕ. Hence a tendency of decreasing α with higher ϕ is observed. On the other hand, we find that the prediction is quite sensitive to the value of magnetostrictive strain ratio β. Note that the above calculation is based on the assumption that β $=1/2$ (i.e., $\lambda_i^{\perp}=-\lambda_i^{\parallel}/2$). If $\beta=1/3$ is adopted for our calculations, a better fit is obtained [see Fig. 1(b)]. In another report,[22] Zhai *et al.*'s magnetostrictive measurement for a pure NFO revealed a field dependent characteristic of the magnetostrictive strain ratio, and its value is actually close to 1/3 for the high field regime. Since the magnetostrictive strain ratio should actually have a very complex response for different materials, our later discussion will adopt two distinct values of β for illustration.

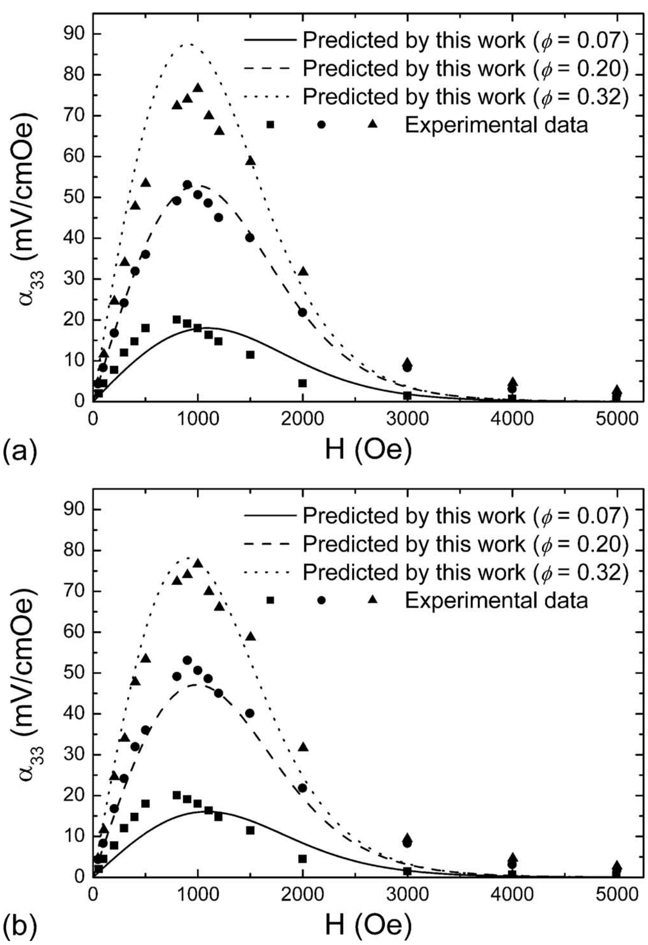

FIG. 1. Comparison of theoretical predictions for the ME coefficient α_{33} of the NFO/PZT 0–3 composites with the experimental data of Zhai *et al.* (see Ref. 16). $\beta(=-\lambda^{\perp}/\lambda^{\parallel})$ is set to (a) 1/2, (b) 1/3.

B. Longitudinal ME response in composites with different β

Figure 2(a) demonstrates the effect of magnetostrictive strain ratio β on the prediction of α_{33} for two distinct values of small ϕ. One can notice that if one adopts a smaller β value (i.e., the response for $\lambda_i^{\parallel}$ is much larger than that for $\lambda_i^{\perp}$), the calculated lines in Fig. 2(a) shift to lower values and this offset seems to be more apparent for higher ϕ. Recalling that the effect of inclusion deformation vanishes (i.e., α_ϕ $=0$) when $\beta=1/2$, but not when $\beta=1/3$ and 2/3 adopted for

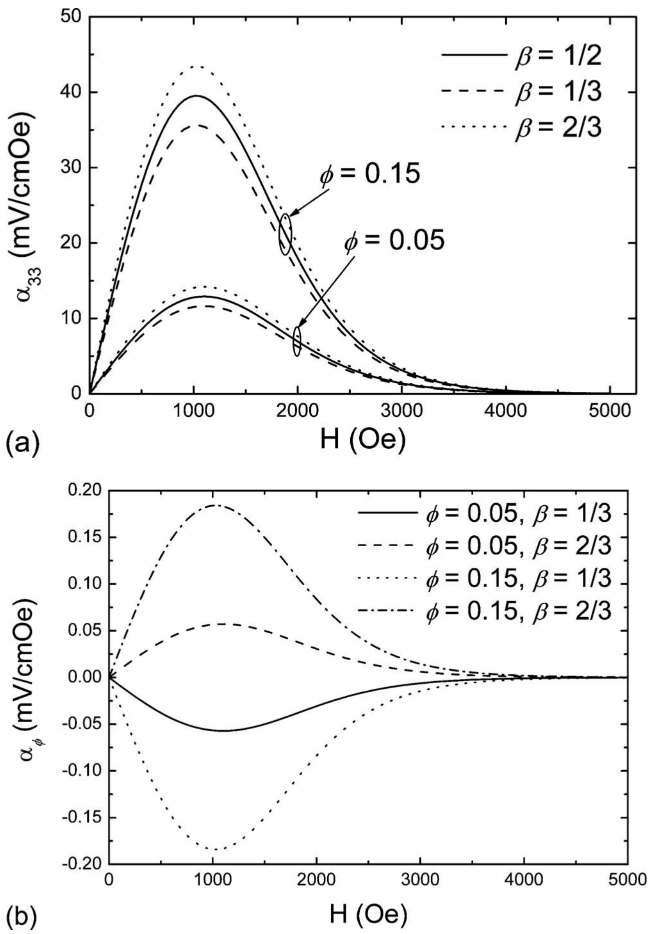

FIG. 2. Variations of the calculated (a) α_{33} coefficient with $\beta(=-\lambda^{\perp}/\lambda^{\parallel})$ and (b) α_ϕ coefficient with β of the NFO/PZT 0–3 composites with ϕ=0.05 and 0.15.

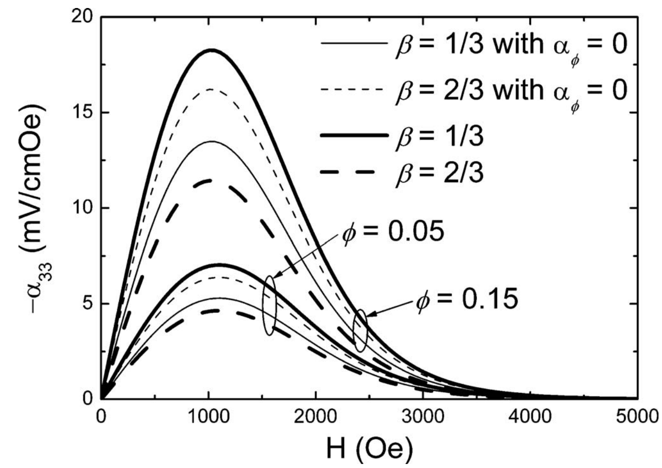

FIG. 3. Theoretical predictions of the ME coefficient α_{33} with/without the effect of inclusion deformation of the NFO/P(VDF-TrFE) 0–3 composites for ϕ=0.05 and 0.15.

illustration in Fig. 2(a). In other words, the calculated results shown in Fig. 2(a) have two contributions, the effects of magnetostrictive strain ratio and the inclusion deformation. It is worth examining the relative contribution of the latter effect in Fig. 2(a). Figure 2(b) shows the calculated results of α_ϕ for $\beta=1/3$ and 2/3 for the two ϕ values. It is seen that the magnitude of α_ϕ for all cases shown is much smaller than the magnitude of α_{33} and indeed does not play a significant role in the calculation of Fig. 2(a). We can conclude that the consideration of inclusion deformation does not have a noticeable effect for the NFO/PZT system. But Fig. 2(b) still reveals that $\alpha_\phi>0$ for the magnetostrictive strain ratio larger than 1/2 and the magnitude of α_ϕ increases with the inclusion volume fraction. This characteristic may be inspected from Eq. (9) by putting $\lambda_{xi}=\lambda_{yi}=\lambda_i^{\perp}=-\beta\lambda_i^{\parallel}$ and $\lambda_{zi}=\lambda_i^{\parallel}$ into Eq. (20).

The above calculations are based on the constituent parameters of the NFO/PZT particulate composite. Next we investigate the inclusion deformation effect on another particulate ME composite system which possesses flexible piezoelectric matrix. Suppose the P(VDF-TrFE) copolymer is used for the piezoelectric matrix material in the ME composite. The constituent parameters adopted for the P(VDF-TrFE) are also listed in Table I. Figure 3 shows the α_{33} coefficient of the dilute NFO/P(VDF-TrFE) composites with magnetostrictive strain ratio $\beta=1/3$ and 2/3. The thicker lines are calculated by the full Eq. (8) and the thinner ones by setting $\alpha_\phi=0$. In the figure, the result for $\beta=2/3$ possesses a larger magnitude than that for $\beta=1/3$. When the α_ϕ term is considered in Eq. (8), the magnitudes of α_{33} for $\beta=1/3$ shift to larger values while the results for $\beta=2/3$ shift to lower values. It is because the sign of α_{33} for NFO/P(VDF-TrFE) is negative, and $\alpha_\phi<0$ when $\beta=1/3$ and $\alpha_\phi>0$ when $\beta=2/3$ as we mentioned previously. In other words, the α_{33} response in an ME composite with $\beta=1/3$ may outperform an ME composite with $\beta=2/3$, if the elastic properties of the constituents facilitate a strong inclusion deformation effect. Similar to our demonstration in Fig. 2(b), the contribution of the α_ϕ term increases with ϕ. One can note from the figure that the change of α_{33} due to the contribution of α_ϕ is significant for ϕ=0.05, comparing to the effect on the NFO/PZT system we discussed in Fig. 2. For ϕ=0.15, the inclusion deformation effect is quite large. It shows that the inclusion deformation can have a vastly different effect on different ME composite materials and is significant for the NFO/P(VDF-TrFE) system. It should not be neglected in the prediction of its ME coefficients. On the other hand, it is thought that other P(VDF-TrFE) based ME composites possessing $\beta<1/2$ in the inclusion may also have similar characteristics of strong inclusion deformation effect such as the popular Terfenol-D, since its reported magnetostrictive responses revealed very small β values ($\approx$0.1) in the medium to high field regime.[23]

From previous figures, we observed that the magnitude of α_{33} is affected by α_ϕ (i.e., effect of inclusion deformation) as well as the value of β (i.e., effect of the magnetostrictive strain ratio) with a significant effect in the NFO/P(VDF-TrFE) system but not so in NFO/PZT. The contribution of β effectively dominates the variation of α_{33} shown in Fig. 2(a). Now we examine the effect of the β value on the α_{33} of the NFO/P(VDF-TrFE) composites. Figure 4(a) shows the calculated α_{33} for ϕ=0.05 and 0.15 with $\beta=1/3$, 1/2, and 2/3. The α_ϕ term in Eq. (8) is set to 0 to disable the inclusion deformation effect, so that the different results solely arise from the variation of magnetostrictive strain ratio β, but not a

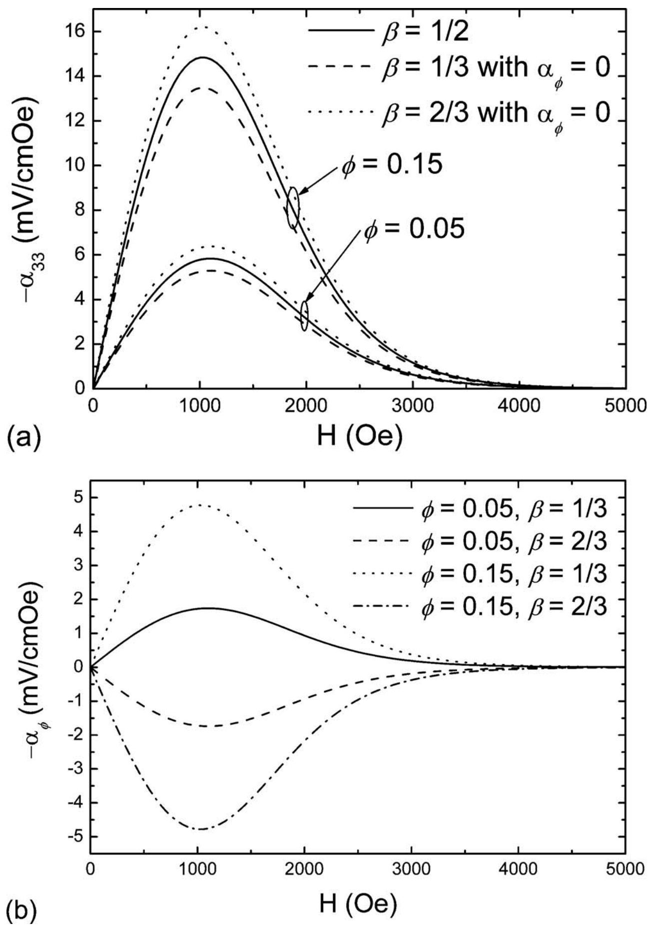

FIG. 4. Variations of the calculated (a) α_{33} coefficient with $\beta(=-\lambda^{\perp}/\lambda^{\parallel})$ with α_{ϕ} set to 0 and (b) α_{ϕ} coefficient with β of the NFO/P(VDF-TrFE) 0–3 composites with ϕ=0.05 and 0.15.

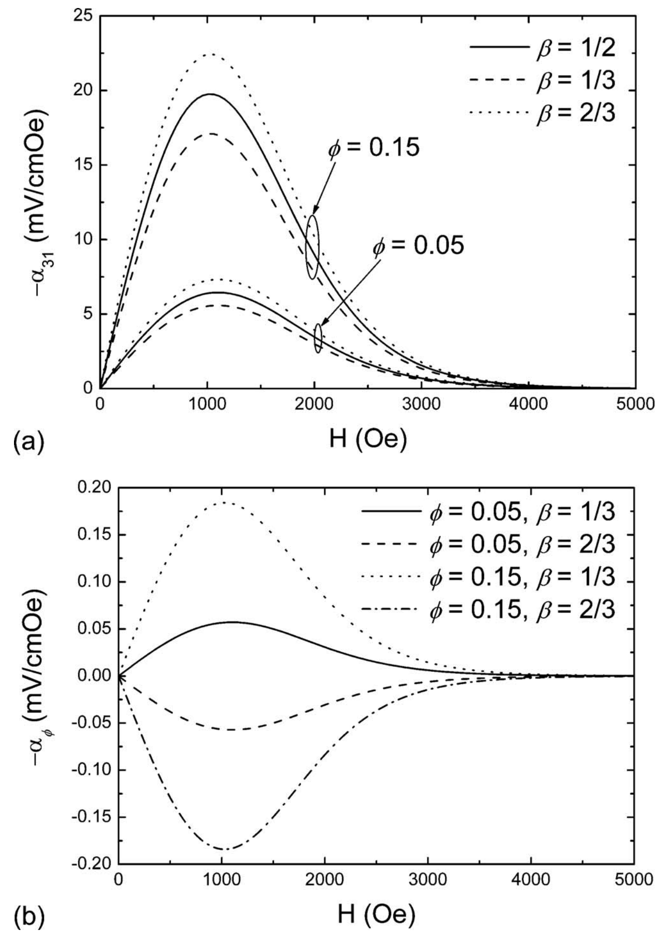

FIG. 5. Variations of the calculated (a) α_{31} coefficient with $\beta(=-\lambda^{\perp}/\lambda^{\parallel})$ and (b) α_{ϕ} coefficient with β of the NFO/PZT 0–3 composites with ϕ=0.05 and 0.15.

coupled effect with $\Delta\phi$. A comparison between Figs. 2(a) and 4(a) shows that the variation of β seems to have a similar percentage change on affecting the α_{33} of the NFO/P(VDF-TrFE) composites, as it does on the NFO/PZT composites. In Fig. 4(b), the curves of α_{ϕ} are shown which have been included in the calculation of the thick lines in Fig. 3. The values of α_{ϕ} shown in the figure are significantly larger than that shown previously [see Fig. 2(b), for the NFO/PZT composites], and the magnitudes are comparable to the α_{33} values in Fig. 4(a). From Figs. 3 and 4, we can conclude that both effects (the variation of β and the α_{ϕ} term) can play a significant role on the modeling of ME properties for the NFO/P(VDF-TrFE) composites.

C. Transverse ME response in composites with different β

The above discussion for magnetoelectricity has been confined to the α_{33} constant. Actually, similar phenomena are also observed in the α_{31} constant. We first examine the transverse ME properties (α_{31}) of the particulate NFO/PZT ME composites. Figures 5(a) and 5(b) show the calculated α_{31} and α_{ϕ} coefficients with ϕ=0.05 and 0.15. The same set of constituent parameters are employed (Table I). As before, setting β (magnetostrictive strain ratio) away from 1/2 introduces the effect of inclusion deformation and displaces the α_{31} curves to higher/lower values. But the inclusion deformation effect apparently has negligible contribution on the resultant α_{31} properties of the NFO/PZT composites [since $|\alpha_{31}|$ shown in Fig. 5(a) is significantly larger than $|\alpha_{\phi}|$ shown in Fig. 5(b)]. On the other hand, the variation of α_{31} in Fig. 5(a) due to the change of magnetostrictive strain ratio seems to have a stronger effect than that for α_{33} [compared with Fig. 2(b)].

Now we consider the α_{31} coefficient of the NFO/P(VDF-TrFE) composites. Similar to that shown in Fig. 3, Fig. 6 shows the calculated α_{31} when the magnetostrictive strain ratio $\beta=1/3$ and 2/3 for the composite with 5% and 15% inclusion volume fractions. Calculated results with and without the effect of inclusion deformation are compared in the figure. It is found that the magnitude of α_{31} decreases (increases) when $\beta=1/3$ (=2/3), an opposite of the feature shown in Fig. 3 for α_{33}. It is because α_{33} and α_{31} have opposite signs, but the α_{ϕ} term always gives the same sign for the same β value, irrespective of whether α_{33} or α_{31} is calculated. In sum, magnetostrictive inclusions with $\beta > 1/2$ in a ME composite is preferred when the α_{31} mode is used. Another point worth noting from Fig. 6 is that the change of α_{31} can be very significant when ϕ=0.15. Normally for a particulate ME composite, $|\alpha_{33}|>|\alpha_{31}|$, thus the contribution due to the same α_{ϕ} is larger for the case of α_{31}.

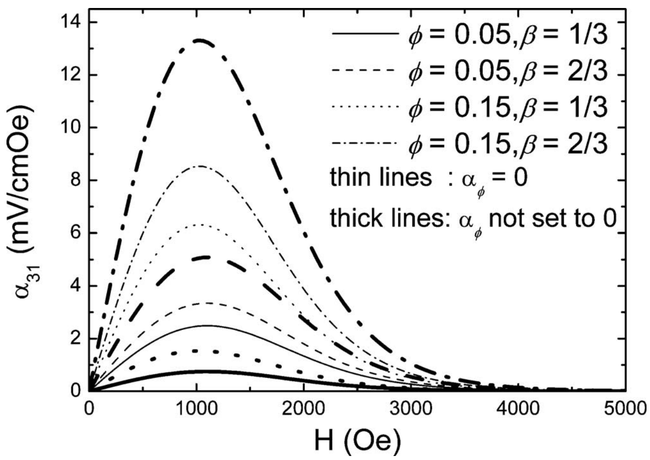

FIG. 6. Theoretical predictions of the ME coefficient α_{31} with/without the effect of inclusion deformation of the NFO/P(VDF-TrFE) 0–3 composites for $\phi=0.05$ and 0.15.

The effect of inclusion deformation has already been found to be significant in the α_{33} mode of the NFO/P(VDF-TrFE) composite (see Fig. 3). It will represent a much stronger factor on affecting the performance of α_{31} coefficient and should be taken into account in theoretical estimations.

In Fig. 7(a), the magnetostrictive strain ratio $\beta=1/3$, 1/2, and 2/3 is examined for the α_{31} coefficient of the NFO/P(VDF-TrFE) composites, under a null α_ϕ term in Eq. (8) [i.e., without the inclusion deformation effect]. This figure shows similar features as in Fig. 4 for the α_{33} coefficient of NFO/P(VDF-TrFE), except that the variations in the magnitude of α_{31} are now much obvious. Figure 7(b) shows the component of α_ϕ contribution in the calculation of Fig. 6. The magnitudes of α_ϕ are identical to that shown in Fig. 4 (for α_{33}). As we can also note this phenomenon from Figs. 2 and 5 for NFO/PZT, this is due to the fact that α_ϕ always give the same values for the same composite sample, irrespective of whether α_{33} or α_{31} is calculated.

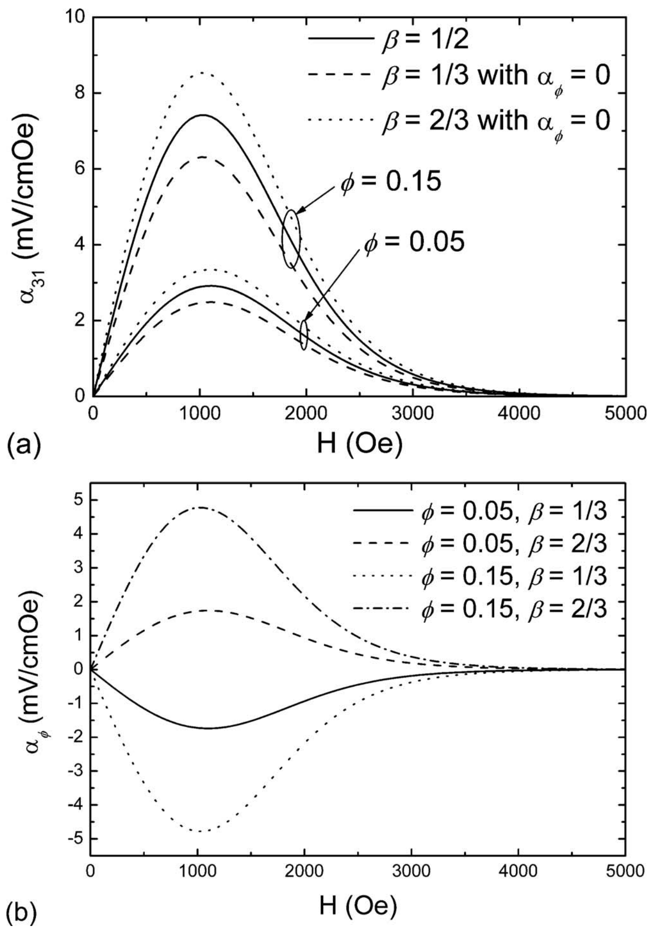

FIG. 7. Variations of the calculated (a) α_{31} coefficient with $\beta(=-\lambda^{\perp}/\lambda^{\parallel})$ with α_ϕ set to 0 and (b) α_ϕ coefficient with β of the NFO/P(VDF-TrFE) 0–3 composites with $\phi=0.05$ and 0.15.

In the above analysis, we have introduced the contribution of $\Delta\phi$ from the deformation of inclusion particles in the calculation of ME properties. The present formulation may be extended to include other contributions, such as the change of the depolarizing factor, by adding an additional term to Eq. (5). In the α_{33} measurement, the inclusion particles are deformed to become spheroids with the depolarizing factor in the poling direction approximately $1/5+2(a/c)^2/15$ where a and c are the radius of the particle along the transverse and longitudinal directions, respectively.[24] Hence the change of depolarizing factor due to the inclusion deformation may be calculated by $(e_{xi}-e_{zi})/15$. However, this change only constitutes about $2.4\times10^{-4}\%$ and $3\times10^{-4}\%$ for the NFO/PZT and NFO/P(VDF-TrFE) systems, respectively. Therefore, we have not included this contribution in the present calculations.

IV. CONCLUSIONS

In conclusion, we have developed a simple analytical model and included the effect of the change of inclusion deformation for the ME properties of magnetostrictive-piezoelectric particulate composites. Analytical expressions have been derived for α_{33} and α_{31} coefficients which characterize the longitudinal and transverse ME responses, respectively. Comparison with the experimental results of NFO/PZT given by Zhai *et al.*[16] shows good agreement. The calculated results suggested that the inclusion deformation effect (α_ϕ) will not be significant for the NFO/PZT composite system. Inclusion deformation effect in magnetoelectricity is severely dictated by the ratio of the transverse and longitudinal magnetostrictive responses in the inclusion (i.e., the β value) as well as the elastic properties of the piezoelectric matrix phase. This effect can be very significant for ME composites with $\beta\gg1/2$ or $\beta\ll1/2$ in the inclusion and flexible piezoelectric matrix. It is found that the NFO/P(VDF-TrFE) composites may be able to possess very large α_ϕ [Eq. (9)] values and the effect should therefore be considered in the selection of materials for designing ME composites and in the estimation of α for similar kinds of ME composites, especially for their α_{31} coefficient.

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