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# Effect of magnetic bias field on magnetoelectric coupling in magnetoelectric composites

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The effect of dc magnetic bias field on the magnetoelectric coupling of a two-component magnetoelectric composite structure is investigated numerically using the finite-element method, in which the nonlinear magnetostress coupling for the magnetostrictive component is taken into account. It is shown that the magnetostress coupling coefficient increases first and then falls down with increasing of the bias field, and this behavior is argued to be responsible for the dependence of magnetoelectric yield on the bias field. The numerical modeling using the ANSYS5.5 finite element algorithm for the  $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$ -epoxy/ $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ -epoxy composite structure gives fairly consistent results with the experiments. © 2003 American Institute of Physics.

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## I. INTRODUCTION

The magnetoelectric (ME) effect is characterized by a variation of the electrical polarization in response to an external magnetic field  $H$ , or an induced magnetization by an external electric field  $E$ . Since the discovery of this effect in ferroelectromagnetic (FEM) compound  $\text{Cr}_2\text{O}_3$  in 1950s,<sup>1</sup> extensive attentions to it have been received, in the earlier time on FEM compounds and recent years mainly on magnetostrictive-piezoelectric composites.<sup>1-4</sup> However, the observed ME effect for most FEMs is too weak to be applicable, while it has been identified that a number of magnetostrictive-piezoelectric composite structures show significant ME output qualified for potential applications.<sup>5</sup> Generally speaking, for the composite structures, the ME effect originates from a product coupling between the magnetostrictive effect from the magnetostrictive phase and piezoelectric effect from the piezoelectric phase in the composites. It is well established that the ME effect is remarkably dependent of the dc magnetic bias field  $H_0$ ,<sup>6-9</sup> onto which an ac magnetic signal  $H$  is imposed. With increasing bias  $H_0$ , the magnetoelectric coupling coefficient  $\alpha_E = (dE/dH)_{H_0}$  increases first and then drops down slowly after reaching the maximum. Nevertheless, the dependence of the ME effect on bias field  $H_0$  is rarely considered in theoretical approaches. For instance, the phenomenological theory proposed recently to explain the ME effect at microwave frequency for lami-

nate piezoelectric/magnetostrictive composites does not take into account of this dependence in a reasonable manner.<sup>10</sup>

A recent research<sup>11</sup> on the magnetoelastic behaviors of Cu-Ni/Cu-Si epitaxial films sheds us a light on understanding the dependence mentioned earlier. It was revealed that the magnetostress coupling coefficient is a nonlinear function of the applied magnetic field and temperature. This allows us to argue that the bias field dependence of ME coefficient  $\alpha_E$  may be ascribed to the nonlinear magnetostress coupling. In fact, it is believed that a bias field favors a parallel spin alignment for all magnetic domains so that the magnetostrictive response is more significant. In this article, we would like to study the dc bias field dependence of the ME effect in a simple two-component composite structure developed in our laboratory.<sup>12</sup> This structure consists of a  $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.9}$  (Terfenol-D)-epoxy mixed component [magnetostrictive component (MSCP)] bonded with a  $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$  (PZT)-epoxy mixed component [piezoelectric component (PECP)], as shown schematically in Fig. 1, where arrows  $M$  and  $P$  show the poling directions of PECP and MSCP, respectively. The experiment was performed by characterizing the longitudinal vibration mode and a very strong ME yield at the resonance frequency was observed. The evaluated  $\alpha_E$  as a function of bias field  $H_0$ , exhibits similar features as other composite systems studied earlier.<sup>6-9</sup> We shall develop a continuum approach by including the nonlinear response of the magnetostress coupling to the bias field. Our finite-element analysis shows a fair consistency between the approach and the experiments, and allows us to argue that an optimized design in terms of the maximal magneto-stress

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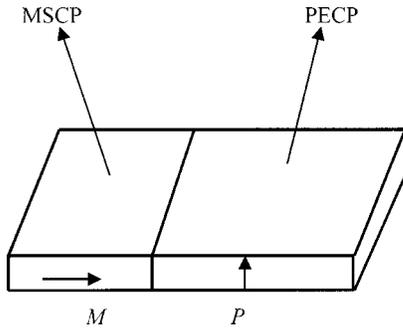


FIG. 1. Schematic illustration of the magnetoelastic composite structure.

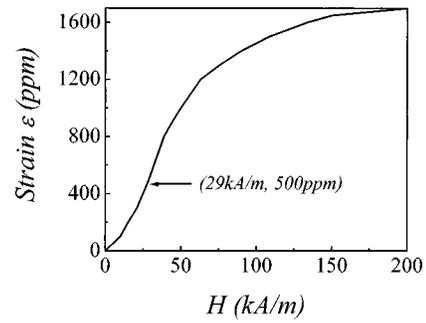


FIG. 3. Measured dependence of the effective magnetostrictive strain on the magnetic bias.

coupling is important for enhancing the ME effect.

### II. NONLINEAR MAGNETOSTRESS COUPLING

In general, the MSCP and PECP as shown in Fig. 1 should be described as bianisotropic media in which the electric displacement and magnetic field vectors depend on the electric field and magnetic induction vectors, respectively.<sup>13</sup> For Terfenol-D, the magnetic field vectors mainly depend on magnetic induction field and the effect of electric field can be ignored here because it is relatively small. Similarly, for PZT, the electric displacement by electric field is considered and the effect of magnetic induction vectors can be ignored. Therefore, the constitution equations for MSCP can be written as<sup>2,14</sup>

$$\sigma = C^H \epsilon - dH, \tag{1}$$

$$B = d^T \epsilon + \mu^\epsilon H, \tag{2}$$

where  $\sigma$  and  $\epsilon$  are the stress and strain tensors;  $B$  and  $H$  are the magnetic flux and field strength;  $C^H$ ,  $d$ , and  $\mu^\epsilon$  are the elastic stiffness constants at a given  $H$ , the magnetostress coupling coefficient, and the magnetic permeability at a given strain, respectively. Here, the induced stress  $\sigma$  as a function of  $H$  is schematically shown in Fig. 2. For an ac magnetic field of amplitude  $\Delta H$ , the corresponding variation of  $\sigma$  is  $\Delta\sigma$ , thus coefficient  $d$  is

$$d = \Delta\sigma / \Delta H. \tag{3}$$

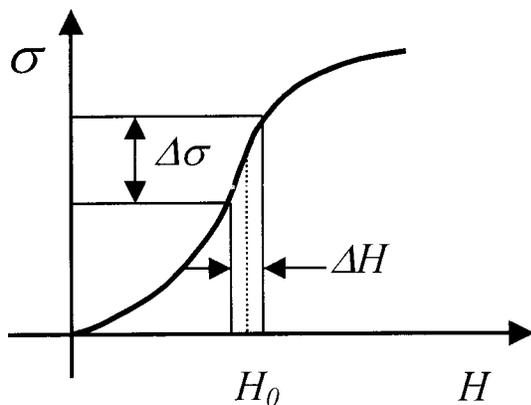


FIG. 2. Schematic illustration of the dependence of stress on the dc magnetic bias field  $H_0$  for the MSCP.

As  $\sigma$  as a function of  $H$  is nonlinear,  $d$  becomes  $H_0$  dependent. The value of  $d$  can be determined by the  $\sigma-H$  relation. This relation is the same as the  $\lambda_s-H$  relation where  $\lambda_s$  is the effective magnetostrictive coefficient and equivalent to strain  $\epsilon_{33}$  along the poling direction.<sup>2</sup> Thus,  $\epsilon_{33}$  must be a function of  $H_0$  too. In Fig. 3 are shown the experimental data on this function.<sup>2</sup> The stress component  $\sigma_{33}$  can be expressed as

$$\sigma_{33} = E_{33} \epsilon_{33}, \tag{4}$$

where  $E_{33}$  is the elastic stiffness constant along the poling direction. Therefore,  $\sigma_{33}$  as a function of  $H_0$  has the same form as  $\epsilon_{33}$ . The curve shown in Fig. 3 may be fitted using a transformation of two hyperbolic curves such as

$$\begin{cases} y = \frac{x}{a + b_1 x} & x \geq 0 \\ y = \frac{x}{a - b_2 x} & x < 0 \end{cases}, \tag{5}$$

where  $a$ ,  $b_1$ , and  $b_2$  are constants to be determined. A simple algorithm leads to

$$\begin{cases} y - y_0 = \frac{x - x_0}{a + b_1(x - x_0)} & x \geq x_0 \\ y - y_0 = \frac{x - x_0}{a - b_2(x - x_0)} & x < x_0 \end{cases}, \tag{6}$$

which is plotted in Fig. 4(b), where  $(x_0, y_0)$  is the coordinates of zero-point defined in Fig. 4(a). Obviously, this point is the inflection point of the curve. The curve excluding the part of  $x < 0$  can be described by

$$\begin{cases} y - y_0 = \frac{x - x_0}{a + b_1(x - x_0)} & x \geq x_0 \\ y - y_0 = \frac{x - x_0}{a - b_2(x - x_0)} & 0 \leq x < x_0 \end{cases} \tag{7}$$

as shown in Fig. 4(c).

We take  $E_{33} = 29$  GPa for MSCP.<sup>12</sup> The inflection point as evaluated from the  $\epsilon_{33}-H_0$  curve is (29 kA/m, 500 ppm), and the largest slope of the curve is 30.3 ppm m/kA. For the  $\sigma_{33}-H_0$  curve (Fig. 4), the inflection point is (29 kA/m, 14.5 MPa) and the largest slope is 878.7 Pa/mA. The parameters

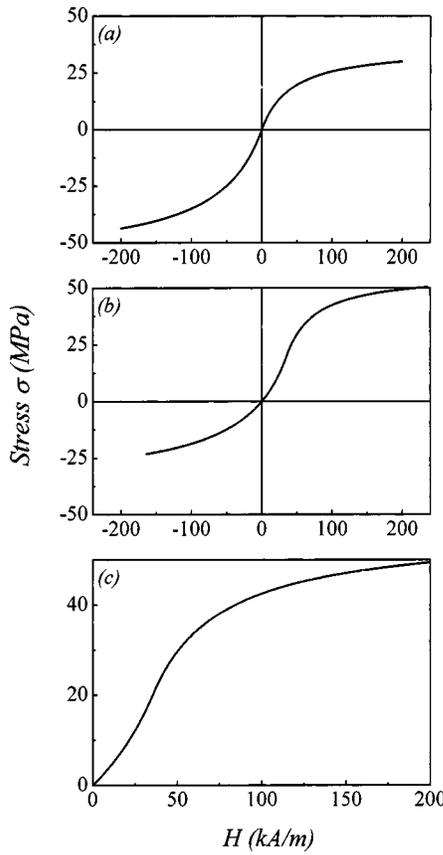


FIG. 4. A transformation of two hyperbolic curves.

in Eq. (7) are  $x_0=29$  kA/m,  $y_0=14.5$  MPa,  $a=1.14 \times 10^{-3}$ ,  $b_1=2.21 \times 10^{-8}$ , and  $b_2=2.95 \times 10^{-8}$ . The evaluated  $\sigma_{33}-H_0$  relation is plotted in Fig. 5.

A differentiation of Eq. (7) yields  $d_{33}$  for MSCP

$$\begin{cases} d_{33}=y' = \frac{a}{[a+b_1(x-x_0)]^2} & x \geq x_0 \\ d_{33}=y' = \frac{a}{[a-b_2(x-x_0)]^2} & 0 \leq x < x_0 \end{cases} \quad (8)$$

The value of  $d_{33}$  at zero field, denoted by  $y'_0$ , is 286.3 Pa/m/A. The normalized coefficient ( $y'/y'_0$ ) as a function of  $H_0$  is

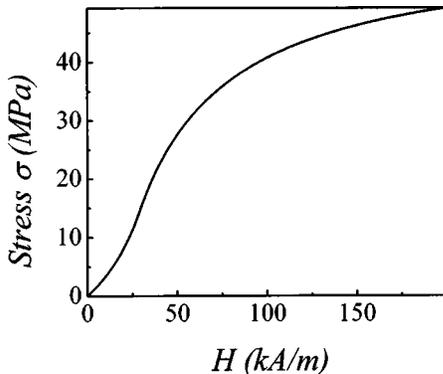


FIG. 5. Calculated magnetostress coupling relationship for the MSCP where the stress is plotted as a function of the magnetic bias field.

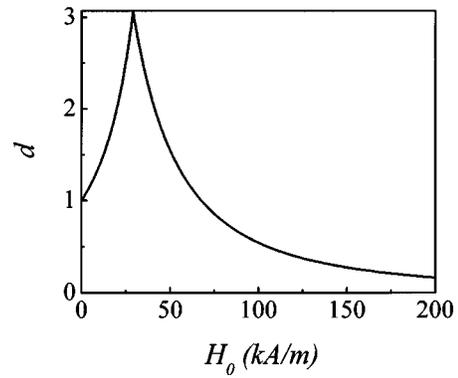


FIG. 6. Calculated magnetostress coupling coefficient  $d$  as a function of the magnetic bias field for the MSCP.

plotted in Fig. 6. Defining  $d_0$  as the magnetostress coupling coefficient at zero bias field, one has the magnetostress coupling coefficient  $d$ :

$$d = d_0 \times d_{33}'/y'_0 \quad (9)$$

From Eq. (9) and Fig. 6 one sees that  $d$  increases rapidly first and then drops slowly with increasing  $H_0$ , as expected earlier.

### III. NUMERICAL MODELING

The effect of the bias field  $H_0$  on the ME effect is calculated using the finite-element method. For the PECP, the constitution equations take the following form:<sup>2</sup>

$$\sigma = C^E \epsilon - e E, \quad (10)$$

$$D = e^T \epsilon + \lambda \epsilon E, \quad (11)$$

where  $\sigma$  and  $\epsilon$  are the stress and strain tensors, respectively;  $D$  and  $E$  are the electric displacement and electric field intensity;  $C^E$ ,  $e$ , and  $\lambda \epsilon$  are the elastic stiffness constants at a given  $E$ , the piezoelectric coefficient, and the dielectric constant at a given strain, respectively. When the MSCP and PECP are bonded together (an ideal bonding interface is assumed here), as shown in Fig. 1, the generation of the ME effect is described as follows: the ac magnetic field applied to the MSCP activates the stress and displacement [Eq. (1)], which is transferred into the PECP via the bonded interface. Consequently, an electric field is generated in the PECP [Eq. (11)]. Obviously, the larger the magnetostress coupling the stronger the magnetostriction for the MSCP, then the higher the voltage output for the PECP.

In the finite-element calculation, a full harmonic response analysis using the software ANSYS5.5/Multiphysics is performed to investigate the dynamic responses of ME coupling to the magnetic bias field. The developed modeling algorithm is described step-by-step in the following.

(1) Finite-element type: a coupled-field solid element is used in the present algorithm.

(2) Material properties: for a realistic calculation on the PECP one needs a complete set of materials parameters for constructing the anisotropic elastic matrix, piezoelectric matrix, and dielectric matrix. The finite-element analysis on the MSCP is performed using the same algorithm as that for the

PECP because the constitutive equations for both components have the same form. Thus, the anisotropic elastic matrix, magnetostrictive matrix, and permeability matrix should be available for the numerical analysis. These parameters provided by the materials manufacturer are listed in the appendix.<sup>15</sup>

(3) Meshing: the sizes of the two plate-like components are 7 mm (length)×6.6 mm (width)×1 mm (thickness), and 8.5 mm (length)×6.6 mm (width)×1 mm (thickness), respectively, for the MSCP and PECP. The interface bonding is achieved by a glue operation in the ANSYS. By means of a prepilot calculation, the numbers of the divided grids are 7 (length)×5 (width)×2 (thickness) for the MSCP, and 10 (length)×5 (width)×2 (thickness) for the PECP. The total elements for the MSCP and PECP are 70 and 100, respectively.

(4) Boundary conditions: in order to meet the requirement of rigid motion there are imposed three zero-displacement conditions along  $z$  axis (thickness), two zero-displacement conditions along  $x$  axis (length), and one zero-displacement condition along  $y$  axis (width). The bottom surface of the PECP and right surface of the MSCP are electrically grounded. The voltage degrees of freedom on the top surface of the PECP are coupled. The ac voltage load is applied onto the left surface of the MSCP to simulate the ac magnetic field on the MSCP, noting that the MSCP is simulated by using the same algorithm as that for the PECP.

(5) The frontal solver technique was used for solving the finite element equations.

(6) The covered range of frequency for the ac magnetic signal is 41 kHz–100 kHz.

#### IV. RESULTS OF NUMERICAL MODELING

The numerical results on the ME coupling coefficient  $\alpha_E$  as a function of bias field  $H_0$  at several frequencies: 41, 69, and 100 kHz, are presented in Figs. 7(a)–7(c), respectively, while the range of  $H_0$  covered experimentally is 0–200 kA/m. It is clearly demonstrated that the bias field  $H_0$  has a significant effect on the ME yield. Given the frequency, the coefficient  $\alpha_E$  increases rapidly first and then drops slowly with increasing  $H_0$ . The effect of  $H_0$  is very significant as the frequency is 69 kHz, while at the other two frequencies, the effect is weaker. In Fig. 7(d) are shown the experimental data on the bias effect.<sup>7</sup> The comparison between the numerical prediction and the experiments indicates a fair consistency between them. Therefore, one is allowed to conclude that the nonlinear response of the magnetostress coupling to the dc magnetic bias field is mainly responsible for the dc bias field dependence of the ME coupling. The numerical algorithm presented earlier seems applicable to this nonlinear behavior in a reasonable way, at least for the bianisotropic MSCP/PECP composite structure as shown in Fig. 1.

The difference in shape between the experiments and numerical prediction is probably ascribed to the following reasons: (1) The nonlinear stress-strain behavior for the MSCP is not included in the calculation. Similar to the magnetostress coupling, the stress-strain relationship may be nonlinear too, i.e., the elastic stiffness constant may be

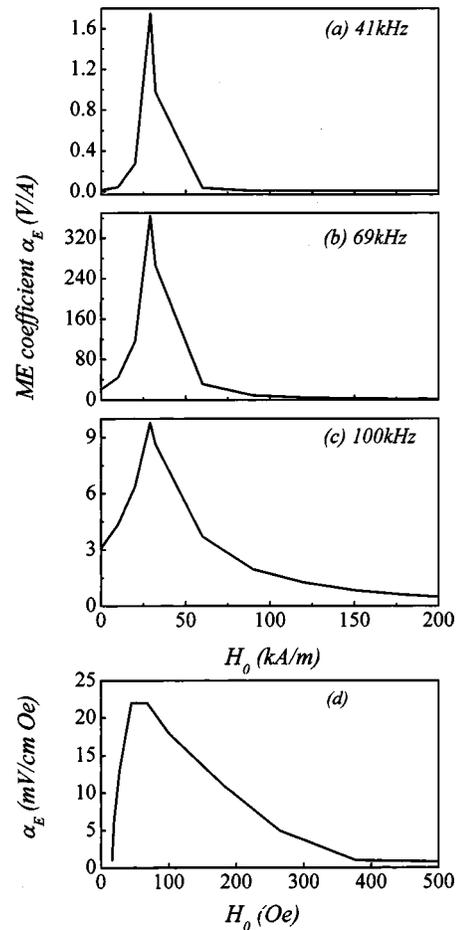


FIG. 7. Numerically calculated magnetolectric coupling coefficient as a function of the magnetic bias field  $H_0$  at different frequencies of the ac magnetic signal: (a) 41, (b) 69, and (c) 100 kHz, respectively; (d) the experimentally measured coupling coefficient  $d$  as a function of  $H_0$  (see Ref. 7).

strain-dependent. This does require a modifying of the magnetostress coupling relationship. (2) The influence of prestress in the materials. It is known that the dc bias field induces a stress in the MSCP. This effect should be taken into account too for a more accurate calculation, although a full consideration of the two reasons mentioned earlier would be challenging. In addition, the influence of temperature and an improved model where the MSCP and PECP are treated as bianisotropic media represent the topics for future study.

#### V. CONCLUSION

In conclusion, the significant effect of dc magnetic bias field on the magnetolectric coupling in a magnetolectric composite structure has been studied using the finite-element method. It has been demonstrated that the dc bias effect of the magnetolectric coupling is ascribed to the nonlinear response of the magnetostress coupling in the magnetostrictive materials to the applied dc bias field. The numerical calculation using the ANSYS5.5 finite element software on the Terfenol-D-epoxy/PZT-epoxy composite structure<sup>12,15</sup> has revealed that the magnetostress coupling coefficient increases rapidly first and then fall slowly down with increasing bias

field, and demonstrated the significant effect of the bias field on the magnetoelectric yield. A rough consistency between the numerical calculation and our experiments has been given.

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## APPENDIX

### A. Materials parameters for the PECP

#### 1. Anisotropic elastic matrix

$$\begin{bmatrix} 7.97 & 3.58 & 3.58 & 0 & 0 & 0 \\ 3.58 & 7.97 & 3.58 & 0 & 0 & 0 \\ 3.58 & 3.58 & 6.68 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1.72 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1.44 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1.44 \end{bmatrix} \times 10^{10} \text{ Pa.}$$

#### 2. Piezoelectric matrix

$$\begin{bmatrix} 0 & 0 & -5.9 \\ 0 & 0 & -5.9 \\ 0 & 0 & 15.2 \\ 0 & 0 & 0 \\ 0 & 10.5 & 0 \\ 10.5 & 0 & 0 \end{bmatrix} \times N/(\text{V m})$$

#### 3. Dielectric matrix

$$\begin{bmatrix} 15.92 & 0 & 0 \\ 0 & 15.92 & 0 \\ 0 & 0 & 15.92 \end{bmatrix} \times 10^{-9} \text{ A(s/V m)}$$

#### 4. Density: 7700 kg/m<sup>3</sup>

### B. Materials parameters for the MSCP

#### 1. Anisotropic elastic matrix

$$\begin{bmatrix} 3.11 & 1.52 & 1.52 & 0 & 0 & 0 \\ 1.52 & 3.56 & 1.52 & 0 & 0 & 0 \\ 1.52 & 1.52 & 3.56 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1.36 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1.36 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1.57 \end{bmatrix} \times 10^{10} \text{ Pa}$$

### 2. Magnetostrictive matrix

$$\begin{bmatrix} 31.3 & 0 & 0 \\ -12.2 & 0 & 0 \\ -12.2 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 21.6 \\ 0 & 21.6 & 0 \end{bmatrix} \times N/(\text{A m})$$

### 3. Permeability matrix

$$\begin{bmatrix} 5.4 & 0 & 0 \\ 0 & 5.4 & 0 \\ 0 & 0 & 5.4 \end{bmatrix} \times 10^{-6} \text{ V(s/A m)}$$

#### 4. Density: 9200 kg/m<sup>3</sup>

- <sup>1</sup>For details of the discovery of magnetoelectric effects in compounds, refer to O. F. de Alcantara Bonfim and G. A. Gehring, *Adv. Phys.* **29**, 731 (1980); M. I. Bichurin, *Ferroelectrics* **204**, 1 (1997).
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