

Effect of magnetic bias field on magnetoelectric coupling in magnetoelectric composites

Y. X. Liu, J. G. Wan, J.-M. Liu, and C. W. Nan

Citation: *J. Appl. Phys.* **94**, 5118 (2003); doi: 10.1063/1.1613811

View online: <http://dx.doi.org/10.1063/1.1613811>

View Table of Contents: <http://jap.aip.org/resource/1/JAPIAU/v94/i8>

Published by the [American Institute of Physics](http://www.aip.org).

Related Articles

Magneto-elastic tuning of ferroelectricity within a magnetoelectric nanowire

Appl. Phys. Lett. **99**, 182901 (2011)

Strain relaxation induced spin reorientation in Fe films on W(110)

Appl. Phys. Lett. **99**, 182501 (2011)

Enhanced magnetoimpedance and field sensitivity in microstructure controlled FeSiCuNbB ribbons

J. Appl. Phys. **110**, 083918 (2011)

Soft-ferromagnetic bulk glassy alloys with large magnetostriction and high glass-forming ability

AIP Advances **1**, 042110 (2011)

Modified Jiles-Atherton-Sablik model for asymmetry in magnetomechanical effect under tensile and compressive stress

J. Appl. Phys. **110**, 063918 (2011)

Additional information on *J. Appl. Phys.*

Journal Homepage: <http://jap.aip.org/>

Journal Information: http://jap.aip.org/about/about_the_journal

Top downloads: http://jap.aip.org/features/most_downloaded

Information for Authors: <http://jap.aip.org/authors>

ADVERTISEMENT

 **AIP**Advances

Submit Now

Explore AIP's new
open-access journal

- Article-level metrics now available
- Join the conversation! Rate & comment on articles

Effect of magnetic bias field on magnetoelectric coupling in magnetoelectric composites

Y. X. Liu

*Department of Civil Engineering, Logistical Engineering University, Chongqing, China
and Laboratory of Solid State Microstructure, Nanjing University, Nanjing 210093, China*

J. G. Wan

Laboratory of Solid State Microstructure, Nanjing University, Nanjing 210093, China

J.-M. Liu^{a)}

*Laboratory of Solid State Microstructure, Nanjing University, Nanjing 210093, China
and Centre for Smart Materials, Hong Kong Polytechnic University, Hong Kong, China*

C. W. Nan

Department of Materials Science and Engineering, Tsinghua University, Beijing, China

(Received 11 March 2003; accepted 5 August 2003)

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.

[DOI: 10.1063/1.1613811]

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 Cr_2O_3 in 1950s,¹ extensive attentions to it have been received, in the earlier time on FEM compounds and recent years mainly on magnetostrictive-piezoelectric composites.¹⁻⁴ 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.⁵ 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 ,⁶⁻⁹ 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.¹⁰

A recent research¹¹ 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 α_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.¹² 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 α_E as a function of bias field H_0 , exhibits similar features as other composite systems studied earlier.⁶⁻⁹ 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

^{a)} Author to whom correspondence should be addressed; electronic mail: liujm@nju.edu.cn

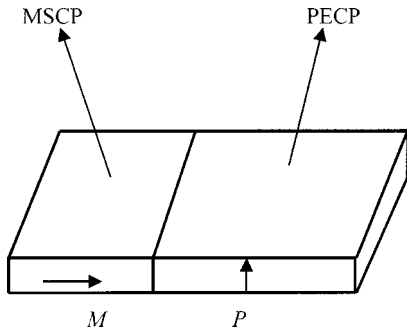


FIG. 1. Schematic illustration of the magnetoelastic composite structure.

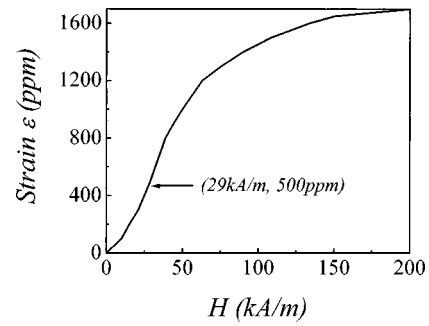


FIG. 3. Measured dependence of the effective magnetostrictive strain on the magnetic bias.

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.¹³ 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^{2,14}

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

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

where σ and ϵ are the stress and strain tensors; B and H are the magnetic flux and field strength; C^H , d , and μ^ϵ 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 σ as a function of H is schematically shown in Fig. 2. For an ac magnetic field of amplitude ΔH , the corresponding variation of σ is $\Delta\sigma$, thus coefficient d is

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

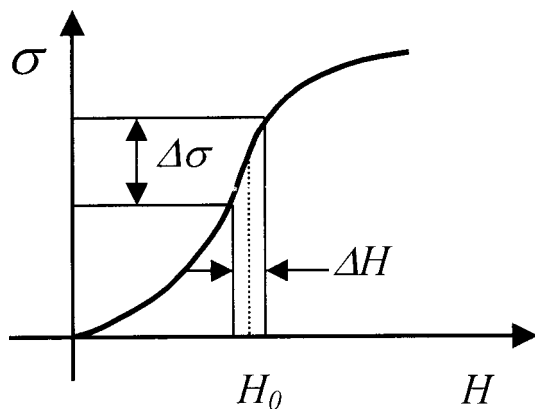


FIG. 2. Schematic illustration of the dependence of stress on the dc magnetic bias field H_0 for the MSCP.

As σ 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 λ_s-H relation where λ_s is the effective magnetostrictive coefficient and equivalent to strain ϵ_{33} along the poling direction.² Thus, ϵ_{33} must be a function of H_0 too. In Fig. 3 are shown the experimental data on this function.² The stress component σ_{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, σ_{33} as a function of H_0 has the same form as ϵ_{33} . The curve shown in Fig. 3 may be fitted using a transformation of two hyperbolic curves such as

$$\begin{cases} y = \frac{x}{a+b_1x} & x \geq 0 \\ y = \frac{x}{a-b_2x} & x < 0 \end{cases}, \tag{5}$$

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.¹² 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

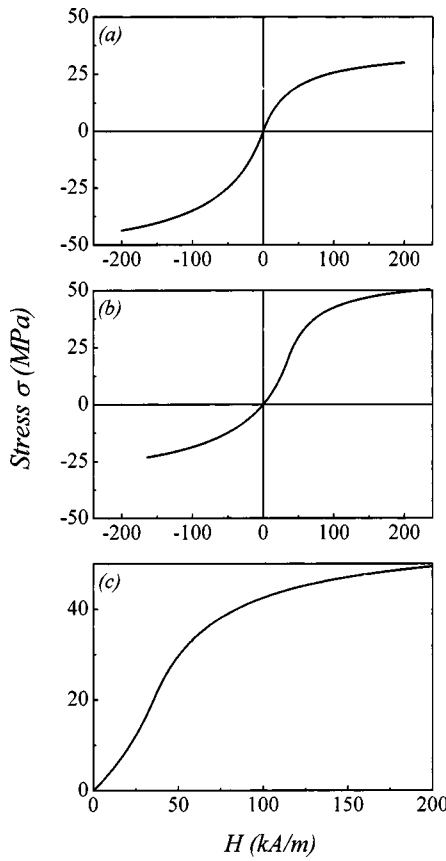


FIG. 4. A transformation of two hyperbolic curves.

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

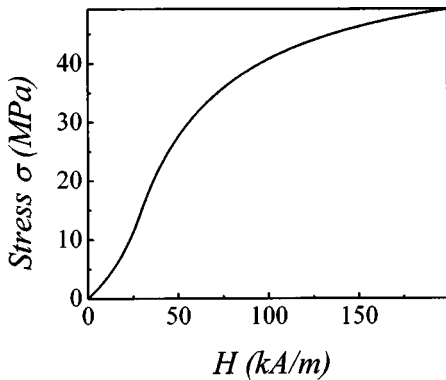


FIG. 5. Calculated magnetostress coupling relationship for the MSCP where the stress is plotted as a function of the magnetic bias field.

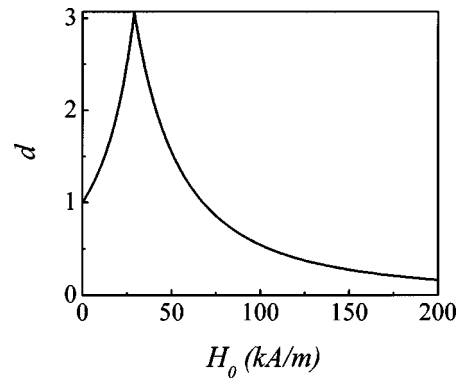


FIG. 6. Calculated magnetostress coupling coefficient d as a function of the magnetic bias field for the MSCP.

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:²

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

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

where σ and ϵ 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.¹⁵

(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 α_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 α_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.⁷ 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

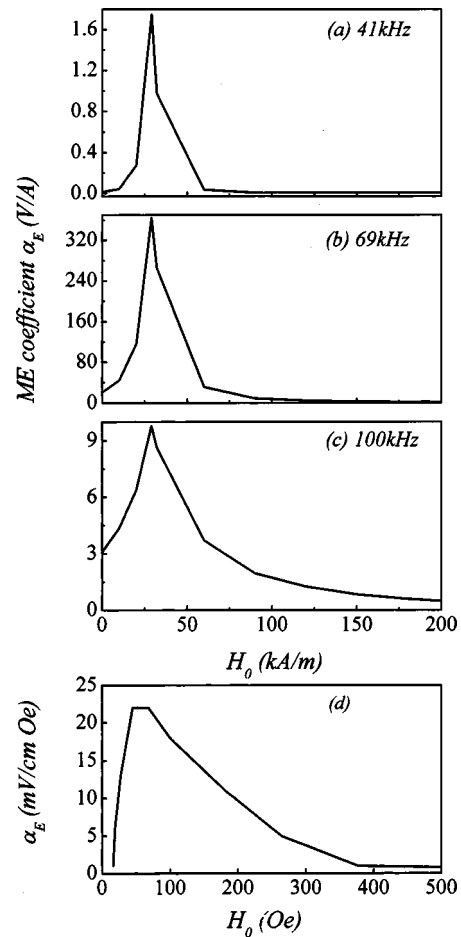


FIG. 7. Numerically calculated magnetolectric coupling coefficient as a function of the magnetic bias field H_0 at different frequencies of the ac magnetic signal: (a) 41, (b) 69, and (c) 100 kHz, respectively; (d) the experimentally measured coupling coefficient d as a function of H_0 (see Ref. 7).

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^{12,15} 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.

ACKNOWLEDGMENTS

The authors would like to acknowledge the financial support from the National Key Research Project for Basic Researches of China (2002CB613303), the National Natural Science Foundation of China, the Center for Smart Materials of Hong Kong Polytechnic University, and NLSSMS of Nanjing University.

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/m³

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/m³

- ¹For 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).
- ²C. W. Nan, *Phys. Rev. B* **50**, 6082 (1994).
- ³C. W. Nan, M. Li, X. Q. Feng, and S. W. Yu, *Appl. Phys. Lett.* **78**, 2527 (2001).
- ⁴G. Srinivasan, E. T. Rasmussen, B. J. Levin, and R. Hayes, *Phys. Rev. B* **65**, 134402 (2002).
- ⁵J. Van Suchtelen, *Philips Res. Rep.* **27**, 28 (1972).
- ⁶G. Harshe, J. P. Dougherty, and R. E. Newnham, *Int. J. Appl. Electromagn. Mater.* **4**, 161 (1993).
- ⁷G. Srinivasan, E. T. Rasmussen, J. Gallegos, R. Srinivasan, Yu. I. Bokhan, and V. M. Laletin, *Phys. Rev. B* **64**, 214408 (2001).
- ⁸J. Ryu, S. Priya, A. V. Carazo, and K. Uchino, *J. Am. Ceram. Soc.* **84**, 2905 (2001).
- ⁹J. Ryu, S. Priya, K. Uchino, and H. E. Kim, *Jpn. J. Appl. Phys., Part 1* **40**, 4948 (2001).
- ¹⁰M. I. Bichurin, I. A. Kornev, V. M. Petrov, A. S. Tatarenko, and Yu. V. Kiliba, *Phys. Rev. B* **64**, 094409 (2001).
- ¹¹M. Ciria, J. I. Arnaudas, L. Benito, C. de la fuente, A. Del Moral, J. K. Ha, and R. C. O'Handley, *Phys. Rev. B* **67**, 024429 (2003).
- ¹²J. G. Wan, J.-M. Liu, H. L. W. Chan, C. L. Choy, G. H. Wang, and C. W. Nan, *J. Appl. Phys.* **93**, 9916 (2003).
- ¹³O. N. Singh and A. Lakhtakia, *Electromagnetic Fields in Unconventional Materials and Structures* (Wiley, New York, 2000).
- ¹⁴M. J. Dapino, R. C. Smith, and A. B. Flatau, *IEEE Trans. Magn.* **36**, 545 (2000).
- ¹⁵B. Charlie, *Sea Technol.* **41**, 17 (2000).