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Estimation of the magnetoelectric coefficient of a piezoelectric-magnetostrictive composite via finite element analysis

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We proposed a new approach for estimating the magnetoelectric coefficient of magnetostrictive/piezoelectric composites via finite element analysis. With this method, the relationship between inputting magnetic field and outputting electric polarization for magnetoelectric composites could be directly calculated. This method offers efficient calculation and is applicable for magnetoelectric composites with any complex structures without restrictions on their connectivity and structures. As examples, the magnetoelectric coefficients of 1-3 type and 0-3 type composites were calculated and the results were found to agree well with literature data.
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I. INTRODUCTION

The magnetoelectric (ME) effect refers to a coupled two-field effect featured by the induction of magnetization upon an applied electric field and/or the induction of electric polarization upon an applied magnetic field.[1] Among a number of compounds and composites with known magnetoelectric effects, ME composites consisting of a magnetostrictive phase (e.g., $CoFe_2O_4$ and Terfenol-D) and a piezoelectric phase (e.g., $PbZr_xTi_{1-x}O_3$ (PZT) and polyvinylidene fluoride) have attracted particular attention due to their relatively strong ME coupling and potential applications in a wide range of electronic components and devices (e.g., sensors and actuations). The performance of a ME composite could be determined by a number of structural factors including contents, distributions and properties of the magnetic and piezoelectric phases.[2,3] Over the years, continuous efforts have been made to establish the relationship between ME property and composite structure through theoretical approaches. In the very early stage of ME composite study, for example, Harshé and Newnham developed an analytical approach to derive the coefficients in various composite systems.[4–6] Srinivasan *et al.* created a model for calculating the coupling effects in laminated magnetostrictive/piezoelectric composites.[7–9] Bichurin and Petrov proposed a generalized effective medium method by introducing an interface coupling parameter k in the calculation, leading to better accuracy of ME calculations,[10] while Dong *et al.* proposed an equivalent circuit model for the calculation of ME coupling in dynamic cases.[11] Nan *et al.* systematically studied the ME coupling effects by analyzing the composites based on Green's function and introduced numerical methods in the calculations.[12–15] However, many of the developed models are only feasible for composites with relatively simple structures but not applicable for composites with a randomly distributed phase; therefore, it would be essential to develop a new approach to calculate the ME coupling coefficient in random systems.

II. THEORY AND MODELLING

In this work, we developed a numerical method for obtaining the ME coefficient in ME composites through finite-element approach. The mechanism for the ME effect has been well documented in the literature, i.e., energy conversion between magnetic and electrical effects is achieved through mechanical coupling and the ME coefficient of a composite, α, can be expressed as

$$\alpha = \frac{\partial P}{\partial H} = k_c \frac{\partial P}{\partial S}\frac{\partial S}{\partial H} = k_c e^p e^m, \tag{1}$$

where H is the externally applied magnetic field, S is the mechanical strain at the interface between the two phases, P is the resulting electrical polarization within the piezoelectric phase, e^m is the piezomagnetic coefficient, e^p is the piezoelectric coefficient, and k_c is the coupling coefficient describing the elastic coupling between the two constituent phases (in an ideal case where $k_c = 1$). In practice, the output voltage (V) is often employed as a measurement for the polarization (P). For a magnetostrictive material subject to external magnetic field H, the strain λ along the magnetic field is $\lambda = dH$, where d is the piezomagnetic strain coefficient. However, in a magnetoelectric composite where the magnetic phases are of multidomains and the magnetic moments orient randomly, such linear relationship would only apply locally. For the whole composite, the strain along a particular direction i can be obtained following Chikazumi's advice,[16] i.e.,

$$\lambda_i = \frac{3}{2}\lambda_s\left[\left(\frac{M_i}{M_s}\right)^2 - \frac{1}{3}\right], \tag{2}$$

where λ_s is the magnetostriction constant, M is the magnetization, and M_s is the saturation magnetization strength with i representing x, y, or z which stands for the magnetization along x, y, or z axis. The negative 1/3 implies that the

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magnetic moments are stochastically oriented in the material when no magnetic field is applied. It should be noted that it is essential to set the calculated value to be zero when it is negative. If we assume that, at the beginning of the magnetization process, the material is pre-stressed such that all magnetic moments are perpendicular to the direction of magnetization, the term 1/3 can then be ignored.[17] In our simulation, the initial strains of magnetostriction parts should be calculated with the formula, and later refined in subsequent simulations. When a ME composite is placed in an external magnetic field, the two phases in the composite should respond according to the magnetostrictive and piezoelectric effect, respectively. When mechanically clamped, the two phases will extrude each other at the phase boundary until they reach a balance. Based on this analysis, our simulation does not only include the two effects described in Eq. (1) but also a self-consistent method in which the magnetic field, deformations and the electric field should be balanced against each other. The self-consistent solving process is described in Fig. 1, expanded from Mudivarthi's sketches.[18,19] The final solution should pass all three converge tests.

We employed, COMSOL (Ver 3.5 a), commercially available software for electromagnetic simulations and calculations. As none of the calculation modules offered in the software functioned directly for ME calculation, attempts were made to combine certain existing modes to simulate the ME effect.[17] As shown in the flowchart presented in Fig. 1, the "multiphysics mode" was employed with several settings/modifications as outlined below:

(1) Three modes were selected in establishing a COMSOL project: (i) "magnetostatics – no current" (in AC/DC module group); (ii) "solid stress-strain – static analysis" (in structure mechanic module group); and (iii) "piezo solid – static analysis" (in piezoelectric effect sub group).
(2) The ME composite was defined as a bulk material placed in air. The whole structure was separated into three subdomains: (i) magnetic phase (activated as "solid stress-strain mode"); (ii) piezoelectric phase (activated as "piezo solid mode"); and (iii) air. The "magnetostatics mode" was active in all domains.
(3) Constituent equations for the magnetostatics mode: the relationship of $\boldsymbol{B}$ vs. $\boldsymbol{H}$ (letters in bold represent vectors) can be expressed as the following two equations: (i) $\boldsymbol{B} = \mu_0\mu_r\boldsymbol{H}$, which is applicable for nonmagnetic phases-air and piezoelectric phases in the composite; and (ii) $\boldsymbol{B} = f(|\boldsymbol{H}|)\boldsymbol{e_H}$, which represents the nonlinear relationship between the magnetization ($\boldsymbol{M}$) and $\boldsymbol{H}$ field in magnetostrictive phase ($\boldsymbol{e_H}$ is the unit vector along $\boldsymbol{H}$ field).
(4) In the "solid stress-strain mode," a number of parameters of the materials (e.g., Young's modulus, Poisson's ratios, densities, and so on) were involved in the computations. Similarly, piezoelectric tensors and mechanic properties were required for the "piezo solid mode" and these were also taken from literature.
(5) "Nonlinear solving method" was assigned for the computation as the problem was nonlinear.

It is important to note that the simulation described above is essentially valid for a static state simulation, which corresponds to the ME effect at low frequency.

III. EXAMPLES AND DISCUSSIONS

Based on the modeling described above, we calculated the ME coefficients for various magnetoelectric composite

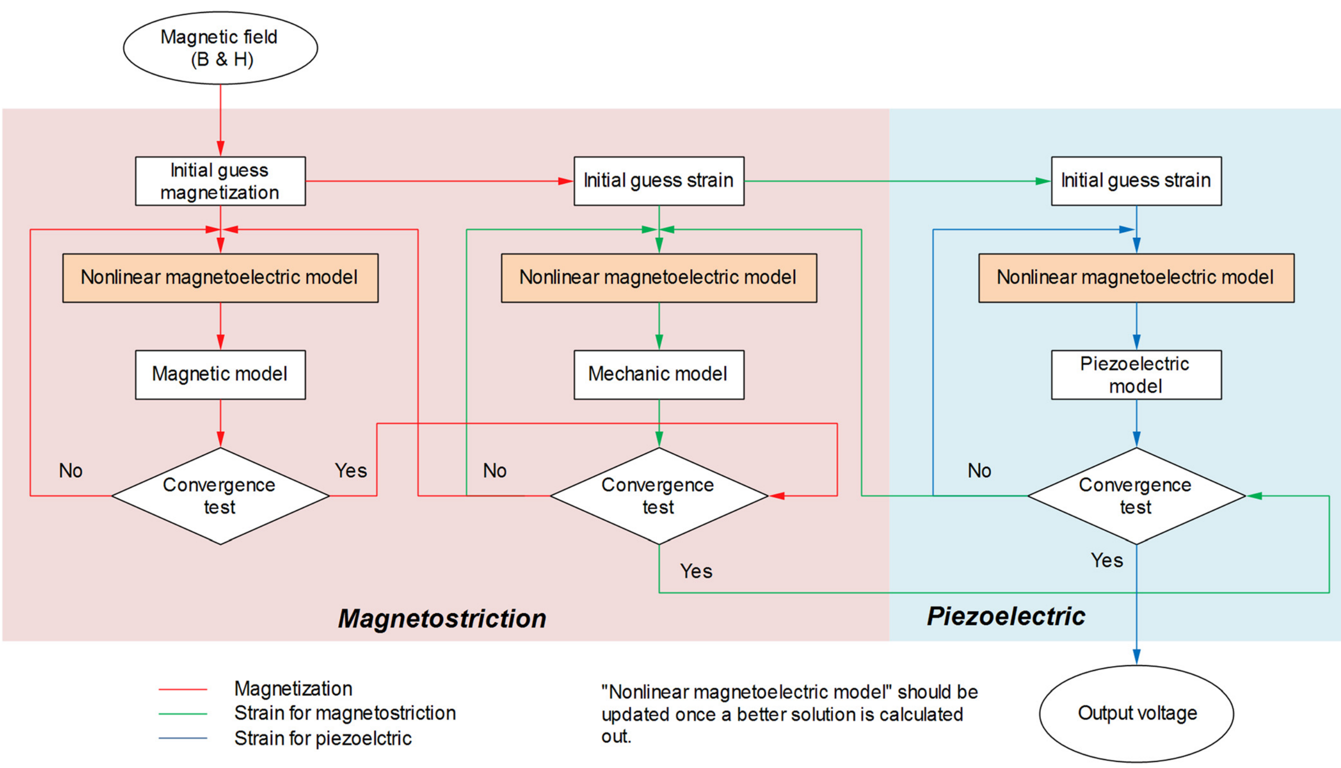

FIG. 1. Self-consistent calculation process.

systems. Here, the results of only two types of structures from our calculations are presented. The first example, as schematically shown in Fig. 2(a), is a 1-3 type ME composite with 9 piezoelectric rod (PZT-5 H) arrays embedded in a magnetic (CFO) cubic matrix. The size of the magnetic cubic was set as $1 \times 1 \times 1$ cm^3, and the distance between the nearest two piezoelectric rods was 0.25 cm. The properties of CFO are set as follows: M_s is $78 \times 5.29 \times 10^3$ A/m (the weight saturation magnetization of bulk CFO is 78 A m^2/kg, and 5.29×10^3 kg/m^3 is the density), λ_s equals to 200×10^{-6}, and the average magnetic susceptibility before saturation equals approximately to 1.3. All PZT-5 H rods were poled along the z axial direction. Mechanical and electric boundaries were set at both the bottom surface and the top surface of the sample. The bottom surface was fixed and grounded, while the top surface was freed for both mechanic and electric boundary conditions. In the simulation, we applied an H field (0–6×10^5 A/m or 0–7540 Oe, where Oe is a more common unit of magnetic field in experiments and 1 Oe = 1000/4π A/m) perpendicular to the top surface and the bottom surface of the composites. A gradient distribution of electric potential was observed from within the piezoelectric rods, signaling the excited electric polarization by H. The averaged electric potential output was estimated by integrating the electric potential on the top surface so that the ME coefficient could be derived. Figs. 2(b) and 2(c) present the electric field output and ME coefficient (dE/dH) upon the application of different magnetic fields. As can be seen, the magnetoelectric response is dependent on the magnetic field. In the low field range, dE/dH increases almost linearly with the increasing H field. Under a high magnetic field, the magnetostriction has become saturated and produced an almost constant electric field in PZT-5 H rod, thereby decreasing dE/dH.

Further information regarding the ME coupling was also obtained through simulation as shown in Figs. 2(d) and 2(e). Figure 2(d) shows the distribution of electric potential within the matrix when H is 3×10^5 A/m. The corresponding displacement of each mass point within the magnetic phase (which performs the level of deformation) is shown in Fig. 2(e). The deformations inside the piezoelectric rods are almost symmetric with the upper half and the lower half pressing against each other. The deformations of magnetic phase are focused near the boundaries inside the cubic and are continuous. Figure 2(f) shows the distribution of magnetization inside the composite (PZT is a non-magnetic medium).

The second example is a 0-3 type composite consisting of PZT-5 H as the continuous phase and CFO particles as the inclusions. The volume fraction of CFO is 12.5%, which can be represented as 125 (this number is variable depending on the phase content) CFO particles (spheres with a radius of 0.062 cm and hence, a volume of ~0.001 cm^3 for each sphere) randomly embedded in a PZT-5 H matrix (cube with edge length of 1 cm). The magnetic field range and the boundary conditions in this composite were identical to those in example 1. It should be noted that unlike the structure in example 1 in which the PZT rods were systematically arranged, the present structure with randomly distributed CFO particles has complicated the simulation process. As it was rather difficult to find a formula to describe "randomness," we decided to employ Monte Carlo method to generate particle coordinates. Fig. 3(a) shows one of the particle distribution cases generated by the random method (note the pink parts refer to CFO this time as opposite to that in Fig. 2). Figs. 3(b) and 3(c) show the averaged E and averaged dE/dH as a function of applied magnetic field. The ME coefficient first increases almost linearly with H until the coefficient reaches a maximum value before descending. It should be noted that Figs. 3(b) and 3(c) are the average values of these 100 groups (details of the averaging method will be provided in the following paragraph). Figs. 3(d)–3(f) present the corresponding electric potential distribution, the deformations along z axis and the magnetization along z axis in the whole structure, respectively.

The averaging method mentioned above was developed with the aim to improve calculation accuracy. As noted from the simulations, the value of ME coefficient of a composite structure may vary with the change of the dispersion "pattern" of the particles (of which the locations are assigned by the Monte Carlo method), even if the total volume

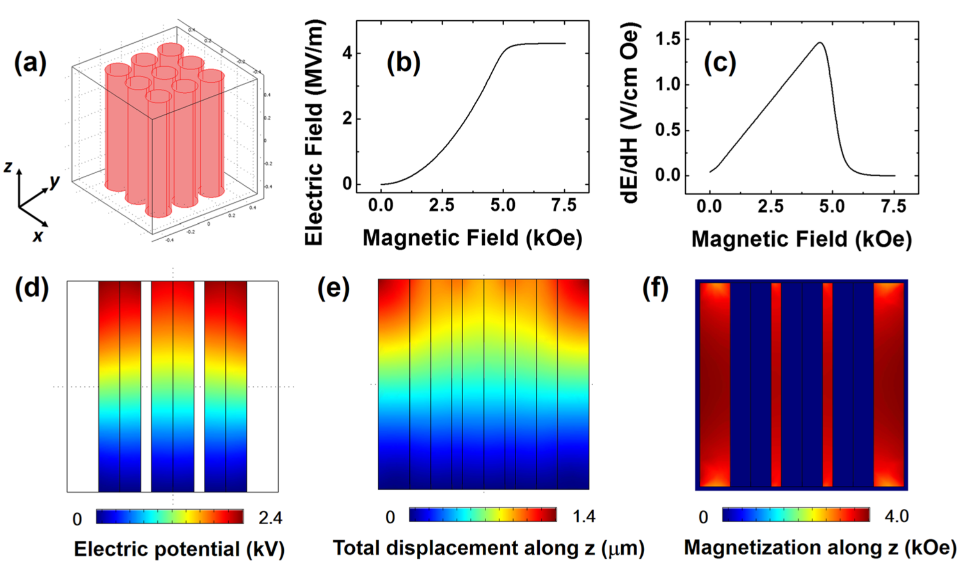

FIG. 2. Simulation for a 1–3 type ME composite: (a) the geometric construction of the structure; (b) outputting electric field vs. inputting magnetic field; (c) dE/dH vs. inputting magnetic field; (d) the distribution of electric potential. The slice is along the center of the structure, the same for (e) and (f), $H = 3 \times 10^5$ A/m; (e) the total displacement along z axis; and (f) the magnetization along z axis.

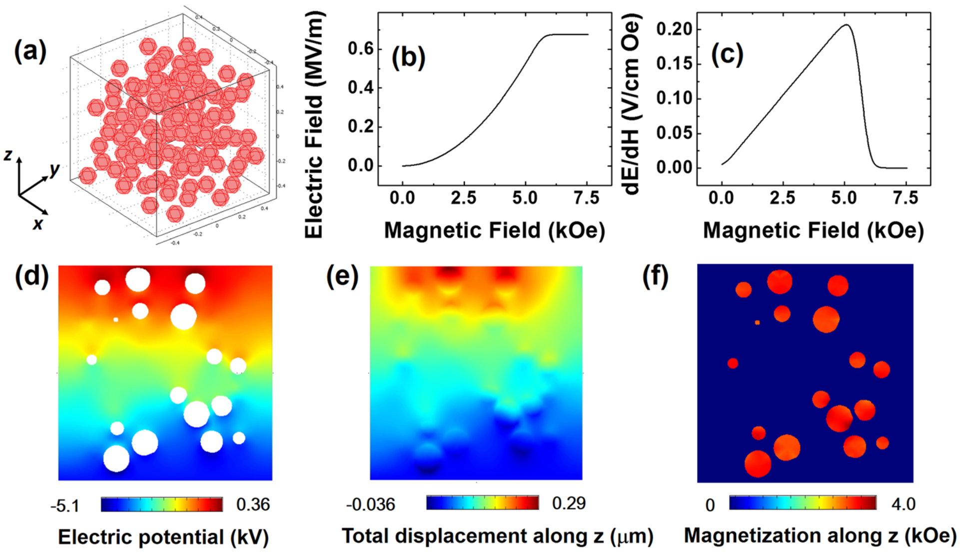

FIG. 3. Simulation for a 0–3 type ME composite: (a) particles distributed in the matrix randomly. This is the distribution pattern found in one of the simulations; (b) outputting electric field vs. inputting magnetic field; (c) dE/dH vs. inputting magnetic field; (d) the distribution of electric potential on center of slice of piezoelectric solid in one of the simulations, the same for (e) and (f), $H = 3 \times 10^5$ A/m; (e) the total displacement along z axis; and (f) the magnetization along z axis.

fraction remains a constant. Theoretically, all possible patterns should be examined in order to obtain an accurate result, i.e., if we first generate dispersion patterns $P_1, P_2, \ldots, P_n$ (by using Monte Carlo method), and find out the ME coefficient for each pattern $\alpha_1, \alpha_2, \ldots, \alpha_n$ (through calculations), then the average value $\alpha_{ave} = (\alpha_1 + \alpha_2 + \ldots + \alpha_n)/n$ could be used to represent the composite only when the value of n is sufficiently large. In our work, we discovered that it would be possible to obtain an acceptable level of accuracy when n $\geq$ 100. Regarding the 0-3 composite discussed above, the values of the ME coefficient for 100 different "patterns" were obtained and were found to follow the Gaussian distribution as shown in Fig. 4. In the calculations, when we set the magnetic field = 1000 Oe, the average value of ME coefficient was 38.4 mV/cm·Oe with a standard error of 0.926 mV/cm·Oe and a larger value of n would lead to an increased sharpness of the peak as shown in Fig. 4; however, a longer computation time is also needed.

We then compared our simulation results with experimental data from the literature and found that they matched

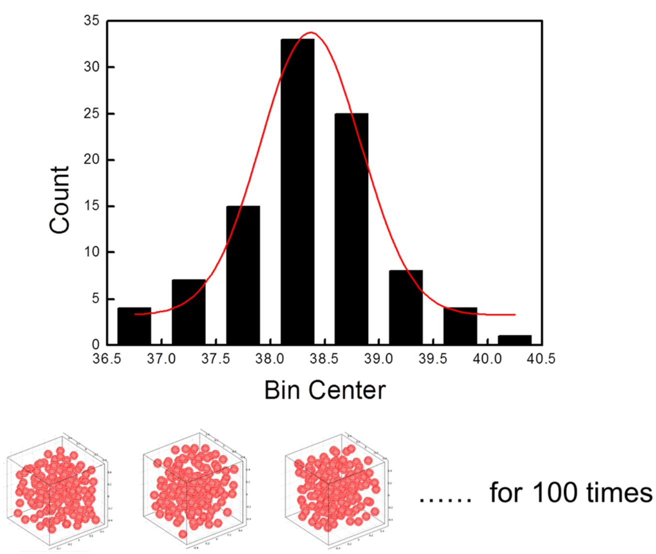

FIG. 4. Distribution of calculated ME coefficient for 100 groups of 0–3 type composite.

well. For example, for the 1-3 type composite with a similar composition and structure as stated in example 1, the ME coefficient in the literature ranged from $\sim 10^2$ to $\sim 10^3$ mV/cm·Oe.[20–22] For 0-3 composites, the magnitude of ME coefficient in the literature falls in the range of tens to a few hundred mV/cm·Oe.[2,23–26] For both structures, the ME coefficient changes with the magnetic field in a way similar to that shown in Figs. 2(c) and 3(c), although the H field for the maximum coefficient may not be identical to that shown in the figures. It is important to note that, in the calculations, the two phases were regarded as an ideal magnetostrictive material (for CFO) and an ideal piezoelectric material (for PZT) without interaction and the distribution of the inclusion was considered to be fully random. However, such assumptions, introduced in the simulations for simplifying the calculations, would not be feasible in real composite materials. In practice, the composite may have various types of "defects" (such as particle aggregations) and the performances of the two phases may be very different from their "standard" properties (e.g., the piezoelectric behavior of PZT may show a nonlinear dependence on the electrical field and time), which may result in deviation of the experimental data from simulation results.[27,28] Theoretical and experimental investigations on the influences of structural "defects" on the ME coefficients are in progress.

IV. CONCLUSIONS

In summary, we developed a self-consistent numerical method to simulate the static response of ME composites. With this method, the relationship between outputting electric field and inputting magnetic can be directly simulated. Two examples with typical 1-3 type and 0-3 type structures were presented in this article to illustrate the simulation. This method is also applicable to simulate the static ME response for composites with any designed structures.

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