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Stress magnetization model for magnetostriction in multiferroic composite

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An alternative to obtain multiferroic materials is the production of composite materials that combine ferroelectric and magnetic materials. In particular, the use of magnetostrictive materials as ferromagnetic phase in composites is very important because the mechanical stress applied in ferroelectric phase induces the appearance of magnetoelectric effect. In this work, we have proposed a generalized model for the magnetostriction dependence with the magnetization of the 0-3 type composite magnetoelectric materials. Including both piezomagnetic and stress dependence in the magnetostriction, a relevant improvement was reached as compared to the ordinary square magnetization model. Based on the Gibbs free energy expansion, the magnetostriction behavior of the composite $(1-x)Pb(Mg_{1/3}Nb_{2/3})-xPbTiO_3/CoFe_2O_4$ at 300 K and 5 K is described. Furthermore, using the piezomagnetic correction, the magnetostriction data for the pure $CoFe_2O_4$ is fitted showing that this ferrite presents a relevant piezomagnetic effect. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4816785]

I. INTRODUCTION

The coupling between magnetic and electric orders can be observed in multiferroic systems, where the magnetoelectric (ME) interaction is defined as the electric polarization of a material in an applied magnetic field or as an induced magnetization in an external electric field.[1] This effect is an important property for development of multifunctional devices, because it is possible to control magnetic and electrical properties using magnetic and electric fields. However, there are few single-phase ME materials, and most of them show very weak ME coupling at room temperature.[2–5] On the other hand, high ME coupling coefficients at room temperature are found in certain class of ferroelectric/ferromagnetic composites materials due to mechanical coupling between the phases.[6,7]

Because of the mechanical stress between the two phases in a multiferroic composite material, many works have been done to understand the effect of the interfacial strain-mediated coupling in these materials.[8–11] One way to obtain a high ME effect is to find a high magnetostrictive material for the ferromagnetic phase. Some materials like cobalt ferrite ($CoFe_2O_4$),[12] Terfenol-D,[12] Cobalt,[12] for instance, present a high values for the relative size changes. In order to understand the ME coupling, a model that describes the response of the magnetostriction is necessary, since the ordinary square model does not describe the magnetostriction of these materials.

In this work, we present a model for the magnetostriction coefficient directly from magnetization measurements. A case study is applied for a cobalt ferrite and a multiferroic 0–3 (particulate) composite in which a ferrite phase is surrounded by a ferroelectric material. In this proposition, we show that the magnetic behavior of these materials is influenced by the piezomagnetic response and the stress of the environment.

II. EXPERIMENTAL DETAILS

The multiferroic sample $(1-x)Pb(Mg_{1/3}Nb_{2/3})-xPbTiO_3$ (PMN-PT) and $CoFe_2O_4$ (CFO) were prepared by the conventional solid state reaction. The polycrystalline ferrite phase (CFO) was prepared using Co_3O_4 and Fe_2O_3 as precursor's powders which were wet mixed 6 ball milled and calcined at 900 °C for 4 h, and grinded at 200 rpm for 10 h. PMN-PT powders were obtained by Columbite method[13] using $MnNbO_4$ as columbite precursor calcined at 1100 °C during 4 h. The columbite precursor were mixed with the other PMN-PT constituents in the correct composition (x = 0.32), calcined at 900 °C, for 4 h, and then grinded at 200 rpm, for 10 h. Finally, the composite powder was prepared by mixing 20% mol of the CFO phase with 80% mol of PMN-PT phase. The mixed composite powders were pressed uniaxially into pellets (~10 mm of diameter and ~10 mm thick), which were densified at 1050 °C, for 0.5 h, through uniaxial hot pressing method (under 6 MPa and O_2 atmosphere). The phase identification of the samples was performed using Rigaku Rotaflex RU200B diffractometer, with CuKα radiation. The apparent density values (ρ_{app}) of the sample were obtained by the immersion method. The theoretical density was calculated considering the proportional average value weight of each constituent magnetic and ferroelectric. A Jeol 5400 LV microscope was used for the scanning electron microscopy analysis of the optically polished and thermally etched sample surfaces. The composites were electrically poled at 25 kV cm^{-1}, for 30 min, at room temperature.[14] The magnetostrictive measurements as a function of applied magnetic field were performed on homemade capacitive cell using a capacitive bridge (Andeen-Hagering model 2500 A) for detecting ΔL/L, from 5 K up to 300 K. Magnetization measurements were carried out using a Physical Properties Measurement System (PPMS) extraction magnetometer by Quantum Design.

III. MAGNETIZATION MODELS FOR MAGNETOSTRICTION

Using the energy approach, in a presence of a magnetic field, the total energy of the material is a combination of the elastic, exchange, and magnetostatic energies.[15] From Gibbs free energy formulation, the total strain of a ferroic material can be expresses by[16]

$$x_{ij} = s_{ijkl}X_{kl} + Q_{ijk}H_k + N_{ijkl}I_kI_l, \tag{1}$$

where s_{ijkl} is a component of the fourth-rank elastic compliance tensor, N_{ijkl} is a fourth-rank tensor for the magnetostriction and the piezomagnetic coefficient, and Q_{ijk} is a third-rank tensor. X_{kl}, H_k, and I_k are the stress, the magnetic field, and the magnetization, respectively. In this case, we have called stress magnetization model (SMM). For an unstressed system ($X_{kl} = 0$) and assuming that the matrix for (Q) is null, Eq. (1) is reduced to

$$x_{ij} = N_{ijkl}I_kI_l. \tag{2}$$

This square magnetization model (SqMM) is consistent with the magnetostriction data for some materials like Ni and Co. However, this model does not take into account materials that present the piezomagnetic effect and are under stress.

To solve this issue, we have proposed a new model that involves both piezomagnetic and stress contribution on Eq. (1). As presented in Eq. (10), the total strain is proportional to the square magnetization corrected by terms which depends on both magnetostriction and piezomagnetic coefficients.

In case of an unstressed condition for a piezomagnetic system, our model here after called unstressed magnetization model (UnSMM) is described by equation

$$x_{ij} = Q_{ijk}H_k + N_{ijkl}I_kI_l. \tag{3}$$

This is the total strain for a piezomagnetic and magnetostrictive material. A more general solution involves all terms in Eq. (1). This solution (Eq. (10)) can be used for a magnetostrictive material inside a non-magnetic media, for example, a 0–3 multiferroic composite. From Eq. (1), the strain is proportional to the stress, to the magnetic field, and to the square magnetization. Here, the magnetization is obtained experimentally and the magnetic field, in the paramagnetic assumption (second term in Eq. (1)) can be replaced by $H_k = \frac{I_k}{\chi_k}$, where χ_k is a component of the magnetic susceptibility. For simplicity, we considered that the applied field is only on the 3-direction. Assuming that the stress is proportional to the grain strain, the relationship between the internal stress and the strain can be expressed, in a first order, as linearly dependent on the strain ($X_{ij} = c_{ijkl}x_{kl}$).[16] From this hypothesis, Eq. (1) is rewrite as

$$x_{ij} = s_{ijkl}c_{ijkl}x_{kl} + Q_{ijk}\frac{I_k}{\chi_k} + N_{ijkl}I_kI_l, \tag{4}$$

where the product $(s_{ijkl}c_{ijkl} = 1)$.[16] Considering a multiferroic magnetoelectric composite as a system with polycrystalline ferromagnetic grains contained in a non-magnetic environment (piezoelectric phase material), the matrix representation of the tensors (Q) and (N) are[16]

$$(Q) = \begin{bmatrix} 0 & 0 & 0 & Q_{14} & Q_{15} & 0 \\ 0 & 0 & 0 & Q_{15} & -Q_{14} & 0 \\ Q_{31} & Q_{31} & Q_{33} & 0 & 0 & 0 \end{bmatrix}, \tag{5}$$

$$(N) = \begin{bmatrix} N_{11} & N_{12} & N_{13} & 0 & 0 & N_{16} \\ N_{12} & N_{11} & N_{13} & 0 & 0 & -N_{16} \\ N_{31} & N_{31} & N_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & N_{44} & N_{45} & 0 \\ 0 & 0 & 0 & -N_{45} & N_{44} & 0 \\ -N_{16} & N_{16} & 0 & 0 & 0 & N_{66} \end{bmatrix}, \tag{6}$$

for the curie ∞/m point group. Using Eqs. (5) and (6) in Eq. (4), a linear equation system can be obtained as

$$\begin{aligned}
0 &= Q_{31}\frac{I_3}{\chi_3} + N_{11}I_1 + N_{12}I_2 + N_{13}I_3 + N_{16}I_6 + x_{22} + x_{33} + 2x_{12} + 2x_{13} + 2x_{23}, \\
0 &= Q_{31}\frac{I_3}{\chi_3} + N_{12}I_1 + N_{11}I_2 + N_{13}I_3 - N_{16}I_6 + x_{11} + x_{33} + 2x_{12} + 2x_{13} + 2x_{23}, \\
0 &= Q_{31}\frac{I_3}{\chi_3} + N_{31}I_1 + N_{31}I_2 + N_{33}I_3 + x_{11} + x_{22} + 2x_{12} + 2x_{13} + 2x_{23}, \\
0 &= Q_{14}\frac{I_1}{\chi_1} + Q_{15}\frac{I_2}{\chi_2} + N_{44}I_4 + N_{45}I_5 + x_{11} + x_{22} + x_{33} + 2x_{12} + 2x_{13} + x_{23}, \\
0 &= Q_{15}\frac{I_1}{\chi_1} - Q_{14}\frac{I_2}{\chi_2} - N_{45}I_4 + N_{44}I_5 + x_{11} + x_{22} + x_{33} + 2x_{12} + x_{13} + 2x_{23}, \\
0 &= -N_{16}I_1 + N_{16}I_2 + N_{66}I_6 + x_{11} + x_{22} + x_{33} + x_{12} + 2x_{13} + 2x_{23}.
\end{aligned} \tag{7}$$

The solution for the total strain along the magnetic field can be found assuming two considerations. First, for a polycrystalline isotropic plated sample, the magnetization is the same for 1 and 2 directions (in-plane, I_{in}). Second, the 3 direction magnetization (out-plane, I_{out}) can be found using $I_s^2 = I_1^2 + I_2^2 + I_3^2$. The expression for the total strain in the 3 direction can be expressed by

$$x_{33} = \left(-\frac{7}{8}N_{31} + \frac{7}{8}N_{33} + \frac{1}{8}N_{11} - \frac{1}{4}N_{13} + \frac{1}{8}N_{66} + \frac{1}{8}N_{12}\right)I_{out}^2 + \left(-\frac{1}{4}Q_{31} + \frac{1}{4}Q_{15} + \frac{7}{8}Q_{33}\right)\frac{I_{out}^2}{\chi_{out}} + \left(\frac{7}{8}N_{31} - \frac{1}{8}N_{66} - \frac{1}{8}N_{11} - \frac{1}{8}N_{12}\right)I_S^2 - \frac{1}{4}N_{44}I_{out}\sqrt{(2I_S^2 - 2I_{out}^2)} - \frac{1}{4}\frac{Q_{15}}{\chi_{out}}I_S^2, \quad (8)$$

where χ_{out} is the out-plane susceptibility and I_S is the saturation magnetization. Using representation matrix for the magnetostriction (N) and piezomagnetic (Q) coefficients, Eq. (1) can be solved for any magnetic particulate system. In order to compare the total strain with an unstressed and piezomagnetic material, a solution for Eq. (3) can be found using the same assumptions. The total strain for UnSMM is

$$x_{33} = (N_{33} - N_{31})I_{out}^2 + (Q_{33})\frac{I_{out}^2}{\chi_{out}} + (N_{31})I_S^2. \quad (9)$$

In Eqs. (8) and (9), the total strain is proportional to the piezomagnetic and magnetostriction terms. This is an expected result for any bulk ferromagnetic material that presents relevant piezomagnetic coefficient. From the total strain, the net magnetostrictive strain (λ) relative to the zero-field state is

$$\lambda = x_{33}(I_{out}, \chi_{out}) - x_{33}(I_r, \chi_r) = \lambda_1(I_{out}^2 - I_r^2) + \lambda_2\left(\frac{I_{out}^2}{\chi_{out}} - \frac{I_r^2}{\chi_r}\right) + \lambda_3\left(\frac{1}{\chi_r} - \frac{1}{\chi_{out}}\right)I_S^2 - \lambda_4\left(I_{out}\sqrt{(2I_S^2 - 2I_{out}^2)} - I_r\sqrt{(2I_S^2 - 2I_r^2)}\right), \quad (10)$$

where I_r is the out-plane remanent magnetization, χ_r is the out-plane remanent susceptibility, and λ_1, λ_2, λ_3 and λ_4 are constants related to (Q) and (N) matrices:

$$\lambda_1 = -\frac{7}{8}N_{31} + \frac{7}{8}N_{33} + \frac{1}{8}N_{11} - \frac{1}{4}N_{13} + \frac{1}{8}N_{66} + \frac{1}{8}N_{12},$$
$$\lambda_2 = -\frac{1}{4}Q_{31} + \frac{1}{4}Q_{15} + \frac{7}{8}Q_{33}, \quad \lambda_3 = \frac{1}{4}Q_{15}, \quad \lambda_4 = \frac{1}{4}N_{44}. \quad (11)$$

For the unstressed and piezomagnetic material, the net magnetostrictive strain (l) relative to the zero-field state is

$$\lambda = x_{33}(I_{out}, \chi_{out}) - x_{33}(I_r, \chi_r) = \lambda_1(I_{out}^2 - I_r^2) + \lambda_2\left(\frac{I_{out}^2}{\chi_{out}} - \frac{I_r^2}{\chi_r}\right), \quad (12)$$

where

$$\lambda_1 = N_{33} - N_{31}, \quad \lambda_2 = Q_{33}. \quad (13)$$

The first term in Eqs. (10) and (12) is related with the magnetostrictive value as found in the literature. The second term in these equations is related with the piezomagnetic contribution of the system. In summary, the magnetostriction can be described by three different approaches: (1) proportional to the square of the magnetization, for unstressed systems and piezomagnetic behavior—(SqMM); (2) unstressed, piezomagnetic and magnetostrictive systems (UnSMM); and (3) stressed, piezomagnetic, and magnetostrictive systems (SMM).

IV. APPLICATION OF MODELS

Figure 1 present the magnetization and normalized magnetostriction measurements of the CFO sample at 300 K. The sample exhibits a high saturation magnetization (80 $\frac{\mathrm{Am}^2}{\mathrm{kg}_{CFO}}$) at 800 $\frac{\mathrm{kA}}{\mathrm{m}}$, when compared with previous results,[17] presenting remanent magnetization and coercive field of 4.8 $\frac{\mathrm{Am}^2}{\mathrm{kg}_{CFO}}$ and 12 $\frac{\mathrm{kA}}{\mathrm{m}}$, respectively. From the magnetization data, we fitted the magnetostriction as function of applied magnetic field using the SqMM and UnSMM models. The results show that there is a good agreement of UnSMM with the experimental data, while the SqMM model only agrees with the experimental data at low field (below 100 $\frac{\mathrm{kA}}{\mathrm{m}}$).

Figure 2 presents the magnetization and the magnetostriction data as a function of applied magnetic field for PMN-PT/CFO composite at 300 K, considering both SqMM

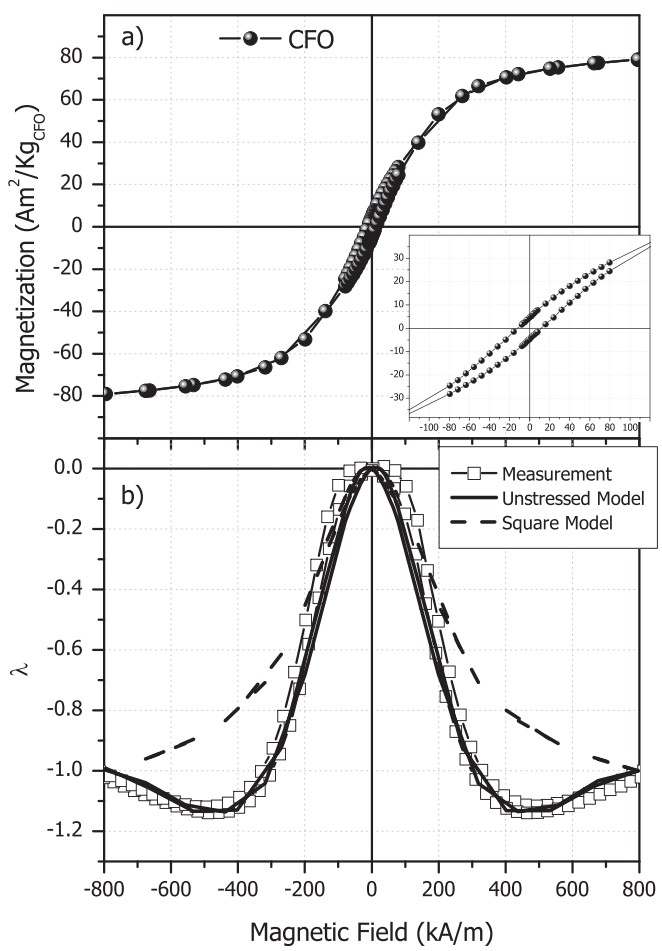

FIG. 1. Magnetization (a) and normalized magnetostriction (b) as a function of applied magnetic field for a cobalt ferrite ceramic at 300 K. The continuous line in (b) refers to the UnSMM and dashed line to the SqMM. Inset in (a) shows details of hysteresis loop. The magnetostriction normalization was obtained at 800 $\frac{\mathrm{kA}}{\mathrm{m}}$.

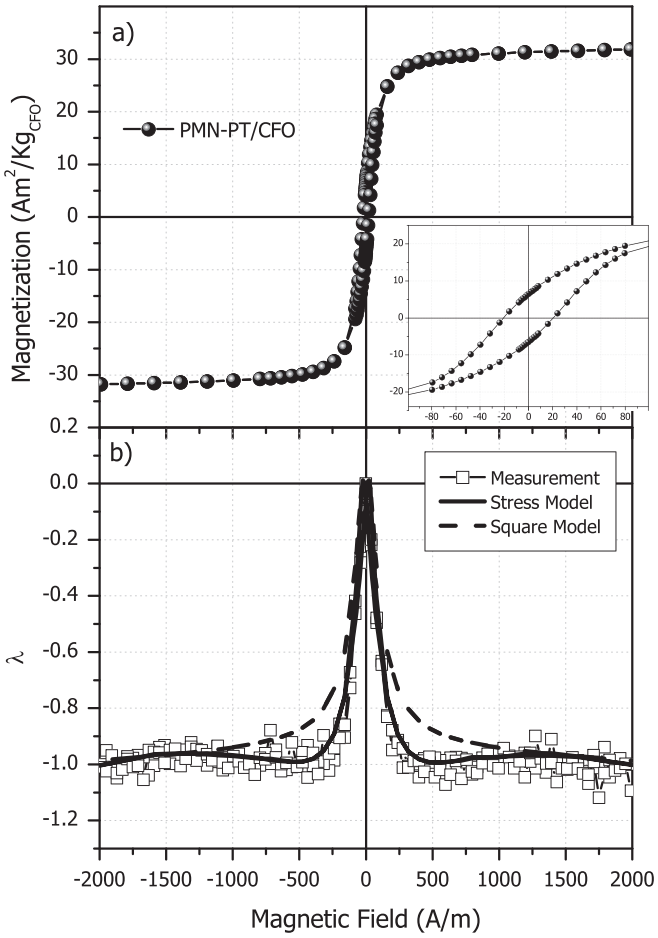

FIG. 2. Magnetization (a) and normalized magnetostriction (b) as a function of applied magnetic field for PMN-PT/CFO at 300 K. The continuous line in (b) refers to the SMM and dashed line to the SqMM. Inset n (a) shows details of hysteresis loop. The magnetostriction normalization was obtained at 2000 $\frac{\text{kA}}{\text{m}}$.

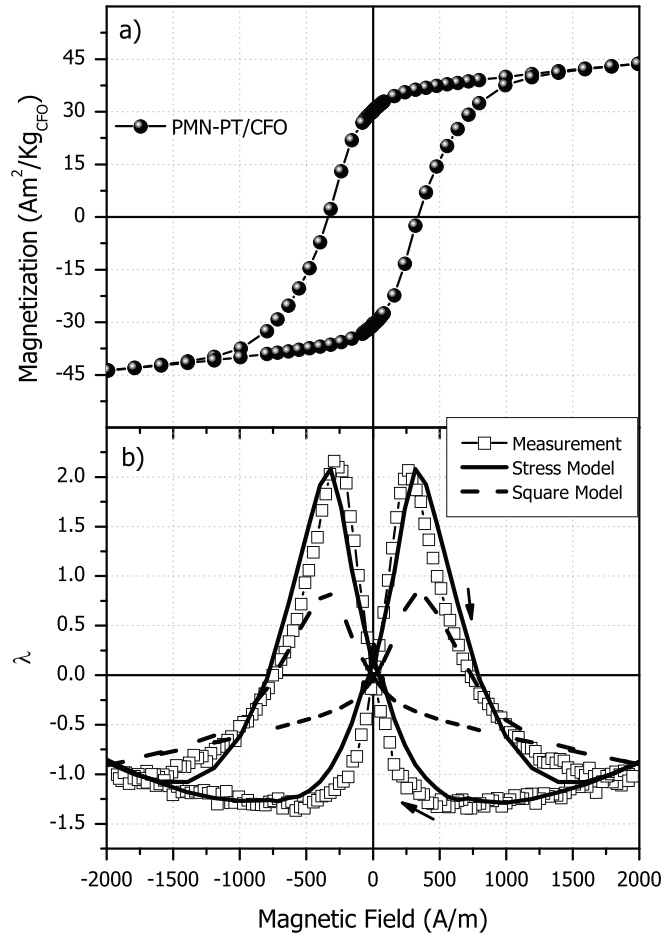

FIG. 3. Magnetization (a) and normalized magnetostriction (b) as a function of applied magnetic field of PMN-PT/CFO sample at 5 K. The continuous line in (b) refers to SMM and dashed line to the SqMM. The magnetostriction normalization was obtained at 2000 $\frac{\text{kA}}{\text{m}}$.

and SMM models. The sample presents a high saturation magnetization of 32 $\frac{\text{Am}^2}{\text{kg}_{CFO}}$ and relatively small remanent magnetization and coercive field, 6.5 $\frac{\text{Am}^2}{\text{kg}_{CFO}}$ and 22 $\frac{\text{kA}}{\text{m}}$, respectively. It is shown that the square model cannot describe the magnetostriction data.

Figure 3 shows the SMM fitting the magnetostriction data at low temperature (5 K). The saturation magnetization reaches 45 $\frac{\text{Am}^2}{\text{kg}_{CFO}}$, as expected higher than that obtained at 300 K. At this temperature, the composite presents a relatively high remanent magnetization and high coercive field, 30 $\frac{\text{Am}^2}{\text{kg}_{CFO}}$ and 340 $\frac{\text{kA}}{\text{m}}$, respectively. The magnetostriction data shows a peak at 400 $\frac{\text{kA}}{\text{m}}$ close to the coercive field.

Table I shows the comparison between the magnetostrictive coefficients found for 3 fittings: CFO at 300 K and PMN-PT/CFO at 300 K and 5 K.

V. DISCUSSION

Comparing Eqs. (10) and (12), the third and fourth terms in Eq. (10) take into account a correction for the magnetostriction. The competition between the expansion (or contraction) of the grain and the contraction (or expansion) of the matrix causes different and opposite stress, Figure 4. The effect of the stress is the change in the dependence of the total magnetostriction with the magnetizaion due to distinct elastics properties of the grain and the matrix.[12] Different values for each constant (λ_1, λ_2, λ_3, λ_4) were found (Table I). As expected,[12] the value for λ_1 and λ_2 at 5 K is greater than the values at 300 K; however, λ_2 present also a different sign at 5 K. At 300 K (Figure 3) and zero field, the ferromagnetic grain is supposed to be unstressed. Considering the coefficient of thermal expansion, during the cooling process, the ferrite (8×10^{-6} K^{-1}) (Ref. 17) will contract more than the PMN-PT matrix (2.4×10^{-6} K^{-1}).[18] This difference between the contractions causes relaxation between the phases, resulting in a less stressed state at low

TABLE I. Comparison between the models (UnSMM and SMM) coefficients for the magnetostriction simulation of CFO and PMN-PT/CFO. λ_1 has the same sign for both models, while λ_2 is different, and these two constants are related with the (N) and (Q) matrix, respectively.

	CFO	PMN-PT/CFO	
	300 K	300 K	5 K
λ_1	-7.15×10^{-8}	-9.55×10^{-8}	-1.66×10^{-7}
λ_2	4.0×10^{-13}	-1.65×10^{-13}	1.09×10^{-12}
λ_3		-1.7×10^{-13}	1.0×10^{-12}
λ_4		1.25×10^{-8}	1.87×10^{-8}

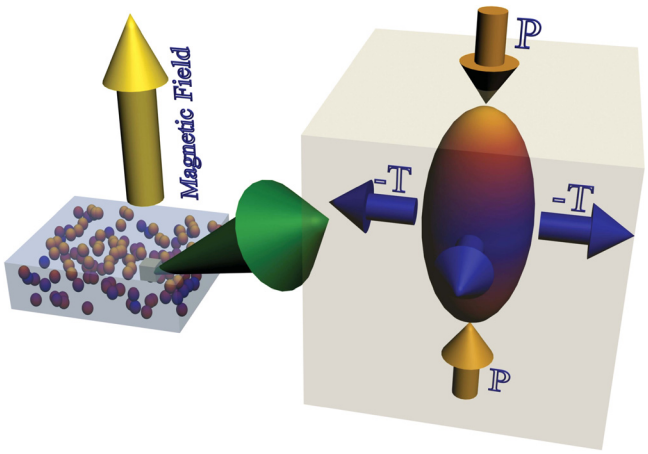

FIG. 4. Schematic cross section of the material showing the matrix stress in the magnetic grain. The effect of the stress on the grain depends on the sign of the magnetostriction: if λ is positive, the grain will push the matrix (pressure, P); if λ is negative, the matrix will pressure the grain (tension, T).

temperature. Because of this, the signs of the constants at low temperature are the same, with equivalent constants for the ferrite at 300 K (Table I).

Furthermore, the magnetostriction effect can be caused by external magnetic field or by stress (stress causes a non-zero magnetization).[15] Because of the stress in the grain due to the matrix, a non-null magnetization causes a different effective magnetic field inside the sample. In a first approximation, the magnetization is proportional to the stress (piezomagnetism). Jiles discussed the effective magnetic field including effects of stress in magnetostrictive materials.[19] In his work, considering the effective magnetic field inside of a ferromagnetic material, Jiles explained the magnetomechanical effect in a material under an external stress σ_0. This new effective magnetic field is proportional to the applied magnetic field, to the molecular field, and to the product of the stress by the magnetostriction derivative. In that model, the stress is kept constant, while for the present model, the stress is proportional to the grain strain. The disagreement for low magnetic field, in the fitting of magnetostriction at 5 K (Fig. 3), is due to both the questionable linear assumption for the susceptibility in the piezomagnetic term and the choice of the Curie point group. For the Curie point group, ∞/m, the material is supposed to be in a polarized state. At high enough magnetic fields, all the magnetic moments align in the direction of the magnetic field (polarized state) and the model fits well the experimental data. At low field, the remanent magnetization determines the symmetry of the magnetic moments similarly to the high field situation. A non polarized state is reached when the magnetic field is inverted, near the coercive field, and the experimental data is not well fitted at low field after magnetic field inversion. Above the saturation magnetic field, the polarized state is recovered and the model again fits the magnetostriction data.

VI. FINAL REMARKS

The simple model represented by Eq. (10) describes with good agreement magnetostriction phenomenon for a magnetic particulate system like the 0-3 multiferroic magnetoelectric composite. In the Gibbs free energy formulation for the total strain, Eq. (1), the assumptions of the stressed grain being proportional to grain strain (Hooke's Law) and the linear relationship between magnetic field and susceptibility are well suited. The correct choice of the magnetic point group is relevant to describe the magnetostriction coefficients as a function of the magnetic field, i.e., knowing the (Q) and (N) matrices, our model describes well the magnetostriction for different types of particulated magnetic systems. Moreover, the proposed model is a contribution to general understanding of the magnetomechanical coupling in magnetoelectric composite materials.

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