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Understanding the dynamic magnetization process for the magnetoelectric effect in multiferroic composites

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Based on a magnetic relaxation model, an approach that includes the spin dynamics is proposed and applied to describe the magnetoelectric (ME) effect frequency dependence for a 0–3 type composite at low temperatures. Our results show that the ME coefficient, in low temperatures, for PMN-PT/CFO ($(1-x)Pb(Mg_{1/3}Nb_{2/3})-xPbTiO_3/CoFe_2O_4$) composite has a step-like behavior on the hysteresis loop for frequency of 1 kHz, contrasting with the results at low frequencies (10 Hz). This approach assumes that the ferromagnetic and ferroelectric phases are coupled through the interactions of the spins of the ferromagnetic phase with the composite phonons by spin/lattice relaxation. *© 2016 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4944889]

I. INTRODUCTION

The multiferroism is characterized by the existence of two or more ferroic orders in the matter.[1–3] In particular, magnetoelectric (ME) multiferroics are the combination of ferromagnetic and ferroelectric orders that present a coupling between the magnetic and electric fields, called magnetoelectricity. This property allows to control the magnetic response due to an applied electric field and vice versa.[4]

The investigation of ME materials has lead to the development of a wide range of devices, such as advanced magnetic sensors, multistate logic, and new magnetic memories for computers.[5,6] However, in single-phase materials, to integrate this phenomenon in devices, it is necessary to find materials with a strong coupling between the ferroic orders. An alternative is to produce multiferroic composites with ferrites and piezoelectric materials, where the mechanical coupling between these leads to the magnetoelectricity.[2] For example, high ME coefficient was obtained for thin films of the composite $PbZr_xTi_{1-x}O_3/CoFe_2O_4$ (PZT/CFO), which reaches 287 mV/cm Oe.[7] Sreenivasulu *et al*. produced an ultrasensitive magnetic field sensor for a sample with PZT fibers and inter-digital-electrodes reaching a peak in the ME coefficient of 250 V/cm Oe at 25 Oe and 47 kHz.[8] In 2014, Lu *et al*.[9] and Chen *et al*.[10] found a zero field ME composite with high hysteresis with respect to the magnetic applied field for the FeCuNbSiB/Ni/PZT (FNP) laminated composite.

Even though many efforts have been done to optimize the ME composites for applications at room temperature, there is a lack of investigations at low temperatures, where one can get relevant information on the nature of the ME coupling. In this direction, most of the low temperature ME results are related to single-phase materials, such as $CaMn_7O_{12}$,[11] $Ba_{0.5}Sr_{1.5}Zn_2Fe_{12}O_{22}$,[12] and $CoCr_2O_4$,[13] and for composites there are some results for the $La_{1-x}Sr_xMnO_3/PZT$.[14,15] Despite the effort to find and characterize these materials,[16,17] several features, such as the magnetoelectric coupling, grain/matrix interface, and grain size effects, are still not well understood.

Another important aspect related to magnetoelectric effect is the dynamic magnetic properties due to spin-spin and spin-orbit coupling. In single phase multiferroics, the magnetoelectricity may rise from different spin coupling processes such as the exchange-striction mechanism, spin-current model, and by the Dzyaloshinskii-Moriya interaction.[18] However, in multiferroic composites, there has been only a few works relating an interaction of the magnetoelectricity with the spins interaction to understand the dynamic ME effect in composite.[19,20] Recently, Yang *et al*. used a phase-field model to predict the ME response of the composite $CoFe_2O_4/BaTiO_3$ based on phase connectivity and phase fraction.[21] Jia *et al*. showed by a theoretical study the influence of the spiral spin density in the ferromagnetic/ferroelectric interface of nanoparticles of Co in $BaTiO_3$ matrix.[22] Bichurin and Petrov investigated the frequency dependency of the bilayer composite PZT/CFO at high frequency in which the maximum ME coefficient was found to be 150 V/cm Oe at 265 kHz.[23] Along these lines, it is important to understand the dynamic magnetic properties of the ferromagnetic phase to comprehend the ME effect in multiferroic composites.

This paper presents a phenomenological interpretation for the ME effect that predicts the ME frequency dependency at low temperature. The approach expands understanding of ME effect to the magnetostrictive response on dynamic magnetization, based on the magnetic relaxation. These concepts are further applied to explain the ME frequency dependence for the composite $(08)(068Pb(Mg_{1/3}Nb_{2/3})-032PbTiO_3)/(02)CoFe_2O_4$ (PMN-PT/CFO) at 5 K.

II. EXPERIMENTAL DETAILS

The multiferroic composite PMN-PT/CFO of 0–3 connectivity was prepared by the conventional solid state reaction following the same procedure as in previously paper.[24] The phase identification of the samples was performed using Rigaku Rotaflex RU200B diffractometer, with CuKα radiation. The

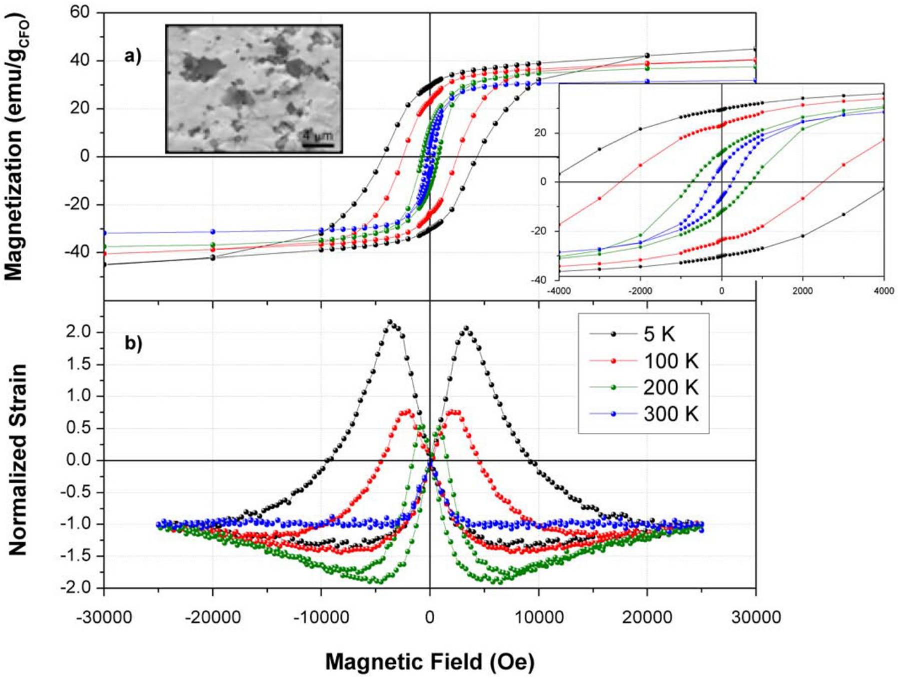

FIG. 1. Magnetic characterization of PMN-PT/CFO as a function of magnetic field at 5 K: (a) the magnetization and the SEM image (inset) with PMN-PT (light) and CFO (dark); (b) the normalized strain.

apparent density values (ρ_{app}) of the sample were obtained by the immersion method. The theoretical density was calculated considering the proportional average weight of each constituent magnetic and ferroelectric component. The composites were electrically poled at 25 kV/cm for 30 min at room temperature.[25]

The magnetostrictive measurements, as a function of applied magnetic field, were performed on a capacitive cell using a capacitive bridge (Andeen-Hagering model 2500A) for detecting the longitudinal strain $\Delta L/L$ at different temperatures. The magnetization and the AC susceptibility measurements were carried out using a Physical Properties Measurement System (PPMS) extraction magnetometer by Quantum Design.

III. RESULTS

Figure 1 shows the magnetization and the strain as a function of applied magnetic field performed at different temperatures for PMN-PT/CFO sample.

Figure 1(a) shows that both the remanent magnetization and the coercive field decreases with increasing temperature (Table I). The detail shows the hysteresis loop in the low magnetic field region. These results can be related to the enhancement of the ferrimagnetic ordering interaction of Co-ferrite compared with the thermal energy. The SEM image (inset) shows two well-defined phases (light for the PMN-PT and dark for the CFO), presenting a distribution of grain size average of about 4 μm for the ferromagnetic phase.

TABLE I. Magnetic properties of PMN-PT/CFO composite at different temperatures.

Temperature (K)	Saturation magnetization (emu/g$_{CFO}$)	Remanent magnetization (emu/g$_{CFO}$)	Coercive field (kOe)
5	48.5	34.0	4.4
100	43.7	26.8	2.6
200	41.4	15.6	0.91
300	34.4	6.9	0.25

Figure 1(b) shows the normalized (at 25 kOe) strain where we observed the same behavior related in the previous paper.[25] The DC magnetic susceptibility (Figure 2(a)) was calculated directly from the magnetization curve derivative $\chi_{DC} = (\frac{\partial M}{\partial H})_T$. The AC susceptibility $\chi_{AC} = (\frac{\partial M}{\partial h_{AC}})_T$ was performed at 10 Hz (Figure 2(a)) and 1 kHz (Figure 2(b)) with AC magnetic field of $h_{AC} = 1$ Oe. While no substantial differences were found comparing Figures 2(b) and 2(c) for the AC susceptibilities at 10 Hz and 1 kHz, the difference between the AC (Figs. 2(b) and 2(c)) and DC (Fig. 2(a)) susceptibilities is non-trivial and can be related to the magnetic relaxation effects.[26–28]

Figure 3 presents the magnetoelectric measurements of the sample of the in-phase induced voltage ($V_{in-phase}$) as a function of the applied magnetic field at different temperatures and frequencies: 10 Hz (Figure 3(a)) and 1000 Hz (Figure 3(b)).

The evolution of $V_{in-phase}$ signal at 10 Hz (Figure 3(a)) in different temperatures shows that the ME voltage present peaks in response to the magnetic field applied. Decreasing the temperature, the coercive field increases (Table I) and, as a consequence, an opening in the $V_{in-phase}$ hysteresis loop is observed. For the $V_{in-phase}$ measured at 1000 Hz and 300 K (Figure 3(b)), the same behavior as the $V_{in-phase}$ measured at 10 Hz was observed. However, the peaks gradually disappear with the decreasing temperature and a step-like behavior is observed (at 5 K), similar to results found for laminated composites.[10]

Figure 4 shows the dependence of the in-phase induced voltage ($V_{in-phase}$) as a function of applied magnetic field at different frequencies at 5 K. The characteristic peaks behavior at 10 Hz disappears for the other frequencies. This effect is a consequence of the dynamic effects on magnetization that contribute on ME voltage behavior, as explained below.

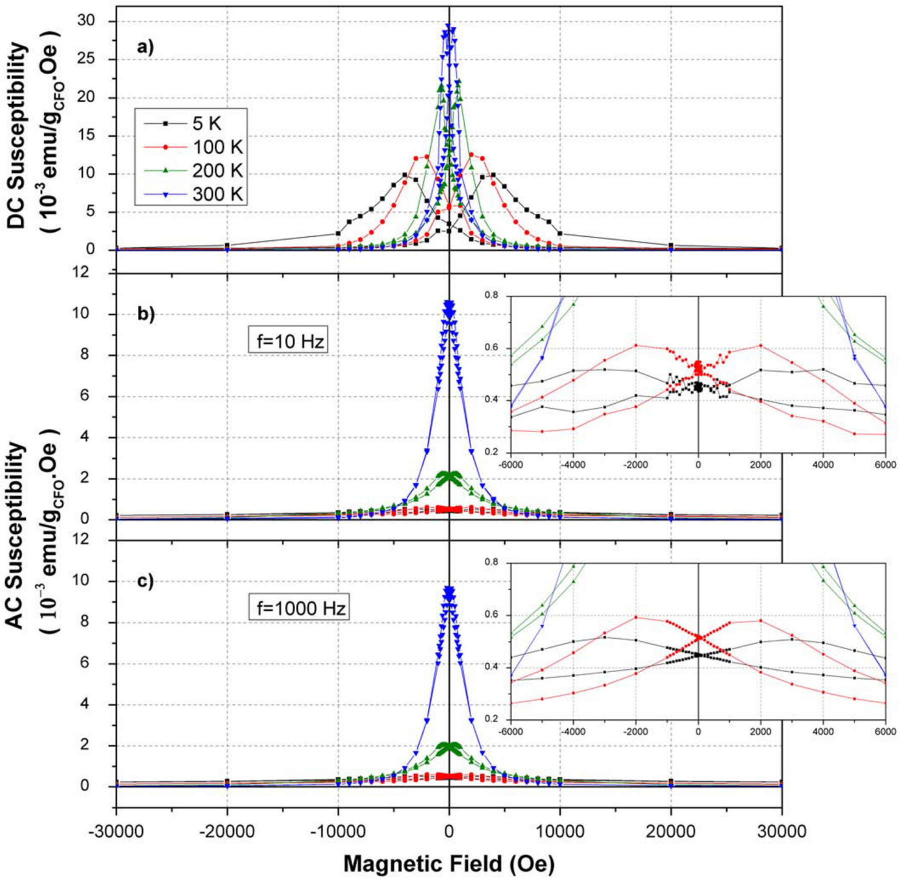

FIG. 2. (a) DC susceptibility (χ_{DC}) calculated from magnetization data. AC susceptibility (χ_{AC}) at frequency of (b) 10 Hz and (c) 1000 Hz with $h_{AC} = 1$ Oe.

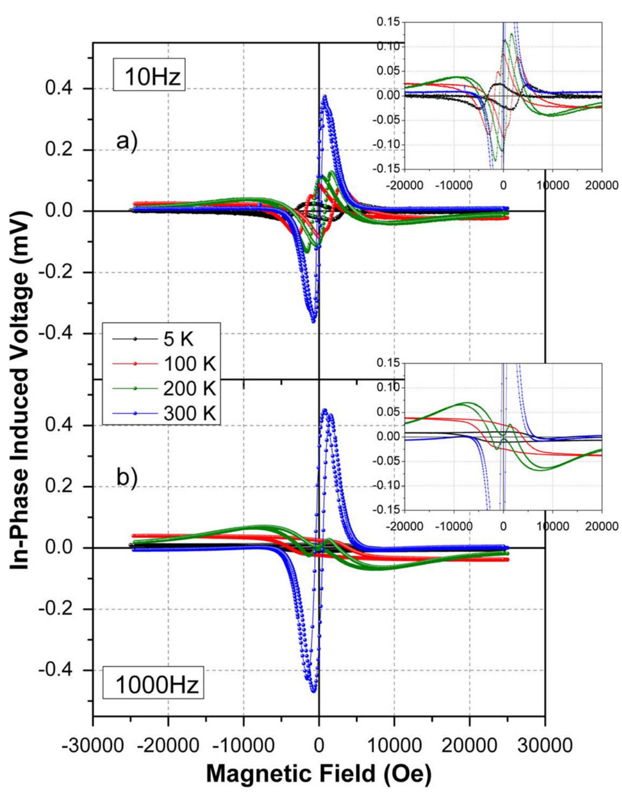

FIG. 3. Magnetoelectric measurements of PMN-PT/CFO at different temperatures and two different frequencies: (a) 10 Hz and (b) 1000 Hz. The measurements were performed with AC magnetic field of $h_{AC} = 1$ Oe. The insets emphasize the low temperature features.

The ME effect is described by a coupling parameter α which is directly proportional to the product between the piezoelectric, ε_e, and magnetostrictive, ε_m, parameters[29]

$$\alpha = \left(\frac{\partial P}{\partial H}\right) = k_c \varepsilon_e \varepsilon_m, \tag{1}$$

where P is the electric polarization, H is the magnetic field, and kc is a coupling factor ($0 \leq k_c \leq 1$). The coefficient ε_e for ferroelectric materials does not depend on the magnetic field; therefore, α will have the same behavior of ε_m, defined as the derivative of the strain with respect to the applied magnetic field.[29,30]

In the magnetostriction measurements, the magnetization is constant in respect to the applied DC magnetic field. Therefore, ME measurements were performed with an AC magnetic field, leading to dissipative effects in magnetic domains. These effects were observed when ME measurements were performed at high frequencies and low temperatures, as shown in Figure 5(b). Our results show a different behavior, indicating that the direct derivative of the strain does not consider the relaxation process due to the AC magnetic field, used in the ME experiments. Thus, it is necessary to include in Eq. (1) the effects of the magnetic relaxation process[26–28] in the magnetostriction effect, as described below.

Recently, our group described for the 0–3 particulate magnetoelectric composites the behavior of the strain due to the application of a magnetic field. It considers terms in first and second order of magnetization (piezomagnetism and magnetostriction) and the stress influence in the

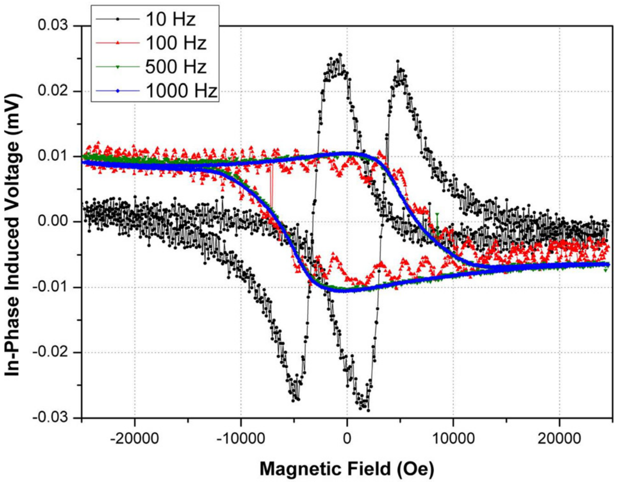

FIG. 4. Magnetoelectric measurements of PMN-PT/CFO at 5 K with different frequencies. The measurement were performed with $h_{AC} = 1$ Oe.

ferromagnetic grains associated with the ferroelectric matrix.[24] In this way, the total strain, λ, of the sample due to the DC magnetic field can be expressed as

$$\lambda = \lambda_1\left(M^2 - M_r^2\right) + \lambda_2\left(\frac{M}{\chi} - \frac{M_r}{\chi_r}\right) + \lambda_3\sqrt{\left(2M_S^2 - 2M^2\right)}(M - M_r) + \lambda_4\left(\frac{\sqrt{\left(2M_S^2 - 2M^2\right)}}{\chi} - \frac{\sqrt{\left(2M_S^2 - 2M_r^2\right)}}{\chi_r}\right). \quad (2)$$

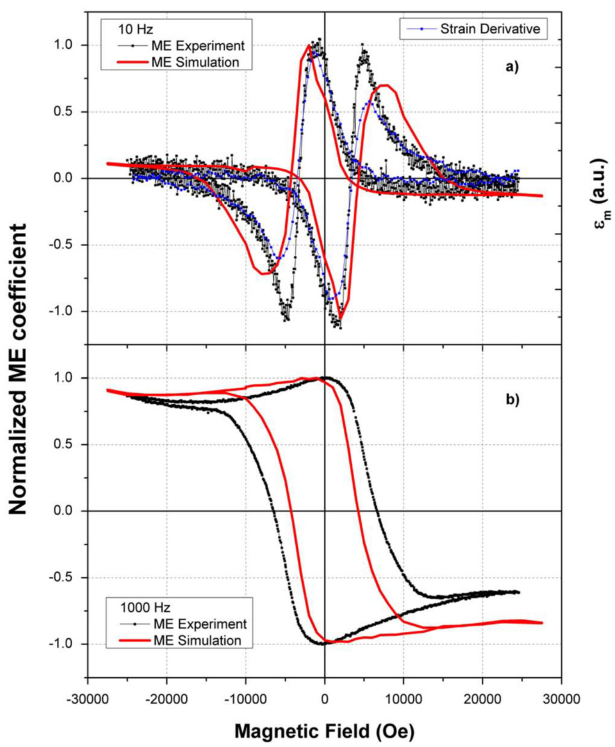

FIG. 5. Magnetoelectric coefficient at 5 K (a) 10 Hz and (b) 1000 Hz and the ME simulation as a function of magnetic field. The measurements were performed with AC field of 1 Oe.

This equation arises when terms of first (piezomagnetism) and second (magnetostriction) orders in magnetization are considered in the equation, which describes the total strain of a ferromagnetic system based in the Gibbs free energy formulation. Furthermore, the effect of stress caused by the ferroelectric matrix in the ferromagnetic grain is also considered, which alters the magnetic properties of the sample. Therefore, $\lambda_1[\mathrm{g/emu}]^2$ and $\lambda_3[\mathrm{g/emu}]^2$ are terms related to the magnetostrictive coefficients (second order); $\lambda_2[1/\mathrm{Oe}]$ and $\lambda_4[1/\mathrm{Oe}]$ are terms related to piezomagnetic coefficients (first order); M, M_r, and M_s are the magnetization, the remanent, and the saturation magnetization; and χ and χ_r are the susceptibility and the remanent susceptibility, respectively. Therefore, ε_m can be written as

$$\varepsilon_m = \frac{d\lambda(M)}{dH} = \frac{d\lambda}{dM}\frac{dM}{dH} = \left(2\lambda_1 M + \frac{\lambda_2}{\chi} - \frac{2\lambda_3 M(M - M_r)}{\sqrt{\left(2M_S^2 - 2M^2\right)}} + \lambda_3\sqrt{\left(2M_S^2 - 2M^2\right)} - \frac{2\lambda_4 M}{\chi\sqrt{\left(2M_S^2 - 2M^2\right)}}\right)\frac{dM}{dH}, \quad (3)$$

where dM/dH is the magnetic susceptibility χ. However, fitting the magnetoelectric effect from Eq. (3) requires not only the dependence for the magnetization and the magnetic susceptibility as a function of applied magnetic field but also the dependence of these properties with the frequency of the AC magnetic field.

To consider the dynamic effects of the magnetization mechanism in the magnetostriction process, we have assumed that for a multiferroic composite material the phonons and spins of the ferromagnetic phase can be treated separately, despite the fact that they are connected through spin/lattice relaxation (Figure 3). Ahlawat *et al.*[31] recently showed that for multiferroic composites, the phonon spectra are the combination of the ferromagnetic and ferroelectric phase phonons. Moreover, any changes in the ferromagnetic phonons configuration, due to the spin/lattice coupling, can induce a renormalization of the sample lattice phonon frequencies.[32] Thus, the magnetic energy due to an external process (AC magnetic field) first acts in the spin system of the ferromagnetic phase. This energy is transferred to the lattice that reaches a thermal equilibrium over a sufficiently long time. As a consequence of the spin/lattice coupling of ferromagnetic phase, there are two important results. If $w\tau \gg 1$ (where w is the frequency of the AC magnetic field and τ is the spin/lattice relaxation time), the temperature of the spin system (T_s) will increase, while the temperature of the lattice (T_l) does not change (the spin systems do not exchange energy with lattice), leading to an adiabatic process, and as a consequence, the response of the magnetic adiabatic susceptibility $\chi_S = (\frac{\partial M}{\partial H})_S$. On the other hand, if $w\tau \ll 1$, the spin-lattice system reaches a thermal equilibrium, characterized by an isothermal susceptibility. This new equilibrium temperature for the ferromagnetic lattice causes a change on the composite phonon spectrum, in agreement to the observed by Ahlawat *et al.*[31]

As a proposal to include these dynamical processes of magnetization in the magnetostriction model represented by Eq. (3), we considered that the measured magnetization M_m is a result of the contribution of spin/lattice (M_{SL}) and non-spin/lattice (M_{NSL}) magnetizations, which can be represented by

$$M_m = M_{SL} + M_{NSL}. \tag{4}$$

In this sense, knowing that the magnetostriction effect is a consequence of the spin-orbit coupling,[33] using Eq. (4), the magnetic susceptibility associated to the dynamic magnetostriction, χ_{SL}, will be the difference between the total susceptibility and the non-spin/lattice susceptibility

$$\frac{dM_{SL}}{dH} = \frac{dM_m}{dH} f(w) - \frac{dM_{NSL}}{dH}(1 - f(w) \Rightarrow \chi_{SL} = \chi_m f(w) - \chi_{NSL}(1 - f(w)). \tag{5}$$

The introduced function $f(w) = 1/(1 + w^2\tau^2)$ is a real function in the Cole-Cole model,[34] which relates the dynamic effects for different contributions of the magnetic susceptibility. The term χ_{NSL} cannot be directly measured; therefore, an indirect approach will be followed. By definition, the magnetic adiabatic susceptibility is related to spins that do not exchange energy with the lattice, being expressed as[26]

$$\chi_s = \chi' \frac{1 + w^2\tau^2}{w^2\tau^2} - \frac{\chi_T}{w^2\tau^2}, \tag{6}$$

where χ' is the real part of the AC susceptibility and χ_T is the isothermal susceptibility (χ_{DC}). Assuming $\chi_{NSL} = \chi_s$ and considering the experimental magnetic susceptibility χ_m, using Eqs. (5) and (6), the part of the magnetic susceptibility that contributes to the spin/lattice relaxation and consequently to the dynamic magnetostriction is

$$\chi_{SL} = \frac{\chi_m + \chi_T - \chi'(1 + w^2\tau^2)}{1 + w^2\tau^2}. \tag{7}$$

Analyzing Eq. (7), for w$\tau \ll 1$, the second term tends to χ_T because in this limit $\chi' \to \chi_T$. Thus, the total χ_{SL} will be proportional to the isothermal susceptibility ($\chi_{SL} = \chi_T$). On the other hand, for w$\tau \gg 1$, χ_{SL} will be proportional to the dynamic susceptibility χ' ($\chi_{SL} = \chi'$). Therefore, using Eq. (3) in Eq. (1) and $dM/dH = \chi_{SL}$, the ME effect can be written as

$$\alpha = k_c\varepsilon_e\left(2\lambda_1 M + \frac{\lambda_2}{\left(\frac{\chi_m + \chi_T - \chi'(1 + w^2\tau^2)}{1 + w^2\tau^2}\right)} - \frac{2\lambda_3 M(M - M_r)}{\sqrt{(2M_S^2 - 2M^2)}} + \lambda_3\sqrt{(2M_S^2 - 2M^2)} - \frac{2\lambda_4 M}{\left(\frac{\chi_m + \chi_T - \chi'(1 + w^2\tau^2)}{1 + w^2\tau^2}\right)\sqrt{(2M_S^2 - 2M^2)}}\right) \cdot \left(\frac{\chi_m + \chi_T - \chi'(1 + w^2\tau^2)}{1 + w^2\tau^2}\right). \tag{8}$$

Equation (8) considers the dynamic effects in the magnetostriction attributing to the magnetic relaxation. Using this equation, the ME effect was simulated for two frequencies, 10 Hz and 1000 Hz, at 5 K. For qualitative purposes, we normalized Eq. (8) by $1/(k_c\varepsilon_e)$ adjusting the simulation scale for better comparison with the experimental results. The constants λ_1, λ_2, λ_3, and λ_4 were obtained from Ref. 24. For the simulation, the only adjustable parameter was the relaxation time, being 23 ms for the best fit. The literature datum for the relaxation time of nanoparticles of CFO are 17 ms at 145 K.[35] The isothermal magnetic susceptibility corresponds to the DC susceptibility, and the χ' is the experimental susceptibility measured at the same frequency as in the ME experiment. These results are shown in Figure 5.

As previously discussed, for w$\tau \ll 1$, the term proportional to χ_T will be dominant in the susceptibility (Eq. (7)). In this case, the ME effect will have the same behavior as the derivative of the strain with respect to the applied magnetic field. The fitting for 10 Hz (Figure 5(a)) shows a good agreement with the experimental ME effect. On the other hand, for w$\tau \gg 1$, the dominant term will be proportional to χ' (Eq. (7)). The fitting for frequency of 1 kHz is plotted in Figure 5(b). The mismatch between the experimental data and the simulation can be associated with the mismatch in the magnetostriction model. The disagreement for low magnetic field in the fitting of magnetostriction is due to both: the questionable linear assumption for the susceptibility in the piezomagnetic term and the choice of the Curie point group.[24]

IV. DISCUSSION

In a ferroelectric material, the electric polarization below the Curie temperature is due to the lattice vibration modes that are non-center symmetric: if one particular mode lowers the crystal energy, the ions will shift to stabilize the structure giving rise to the spontaneous polarization. A variation in the electrical polarization of the crystal can be induced altering the equilibrium condition of vibration modes.[36] As previously reported, Ahlawat *et al.*[31] showed the existence of a coupling between the spin of ferromagnetic phase and phonons in magnetoelectric composites mediated by strain interactions. Thus, knowing that the spin ordering affects the phonon frequencies of the lattice,[37] the interaction of the external magnetic field with the ferromagnetic phase changes the phonon frequencies of the composite. This new phonon frequency spectrum

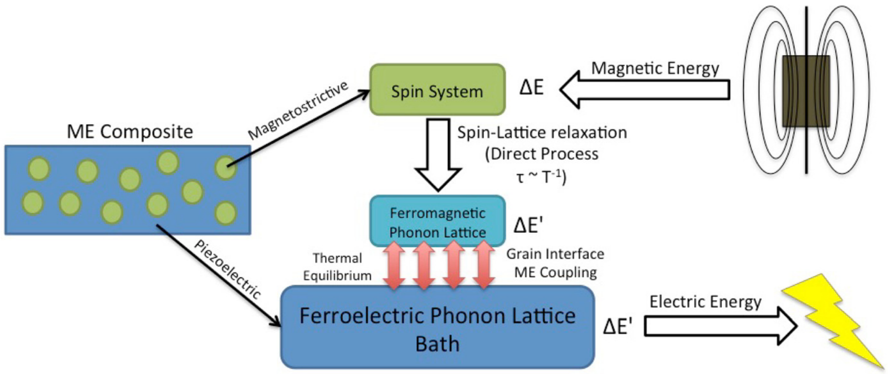

FIG. 6. Illustration showing the ME effect in composites. The spin and phonon systems of the ferromagnetic phase are coupled by the spin/lattice relaxation. The composite phonons spectrum is a result of the coupling between the ferroelectric and ferromagnetic phases.

changes the equilibrium position of the ions in the ferroelectric phase, giving rise to the magnetoelectric effect. Furthermore, based on the molar ratio between the phases in the composite (80% PMN-PT and 20% CFO), the ferroelectric phase can be considered as a thermal reservoir for the ferromagnetic phase (inset of Figure 1(a)). In this way, the magnetoelectric effect in the composite is related with a direct coupling between spins of the ferromagnetic phase and phonons of the composite, similar to the ME effect in single-phase materials.[2,29]

Figure 6 is a schematic diagram illustrating the phonon coupling between ferromagnetic and ferroelectric phases.

The magnetostriction phenomenon is an effect of the applied magnetic field on lattice in the form of elastic strain. This is usually characterized by the deformation along the direction of the magnetic field.[33,37] However, being a manifestation of a magnetoelastic interactions, the effect depends on the origin of the crystalline anisotropy and the strength of the spin/orbit coupling of the magnetic ion.[33,37] Furthermore, the spin/orbit coupling is related to the coupling between the orbital momentum (L) and spin angular momentum (S) ($H = \lambda L \cdot S$). Cobalt ferrites are known to present high values for the magnetic anisotropy energy, which is related to the trigonal field (along [111] direction).[38–40] In particular, this trigonal field induces a splitting in the free-ion (Co^{+2}) energy level in the octahedral site of the spinel structure.[38–40] Moreover, the orbital moment is zero for the higher level of this splitting; therefore, the electrons at this level do not contribute to the spin-orbit coupling. In this sense, the non-(spin/lattice) contribution in Eq. (3) is related to electrons in the level that contribute to the total magnetization rather than to the spin/orbit coupling. Due to the small splitting energy, these electrons can be excited either by temperature or by AC magnetic field.[38–40] Thus, the frequency dependence of the magnetoelectric effect is associated to the dynamic population, between the lower and higher levels, in the trigonal field splitting of the cobalt ferrite phase. In this sense, low frequencies of magnetic field at low temperatures enhance the spin-lattice coupling resulting in isothermal coupling between spin and lattice of ferromagnetic grains, and a higher vibrational mechanical energy is transferred to the ferroelectric reservoir. On the other hand, the increase of magnetic field frequency changes the populations of energy levels of Co-ferrite reducing the spin-lattice coupling consequently reducing the vibrational energy in the ferroelectric phase reservoir. In this condition, higher orders of the magnetostriction effect can be disregarded and the behavior of magnetoelectric curve is similar (in first order) to the magnetization curve.

V. CONCLUSION

In conclusion, the ME effect was modeled to predict changes in the ME response of the PMN-PT/CFO with the frequency of the AC magnetic field at 5 K. Using the adiabatic assumption for the magnetic susceptibility, it was proposed that the ferromagnetic and ferroelectric phases are coupled by the interaction of the spins of the ferromagnetic phase with the composite phonons through spin/lattice relaxation. The current approach assumes that the non-(spin/lattice) susceptibility (χ_{NSL}) (spins uncoupled with the lattice) decreases the spin/lattice susceptibility (χ_{SL}) (spins coupled with the lattice) (Eq. (5)). The χ_{NSL} is related to the spins whose angular momentum of the energy level is presumed to be zero. Therefore, the magnetoelectric effect in composites is correlated to the dynamics of the spin/lattice interaction of the ferromagnetic phase. This dynamic effect impacts the modes of oscillation of the composite phonons, causing a displacement of the equilibrium position of the non-center-symmetric ions, inducing a variation of the electric polarization. One of the most relevant aspects in this approach is the fact that the only adjustable parameter is the relaxation time. Advances on the understanding of the dynamic ME effect in composites make it worthwhile to control the ferromagnetic/ferroelectric grain sizes, the grain interfaces, and the microstructure, in order to tailor the magnetic relaxation time. These results may lead to new applications of multiferroic composite based on the dynamic features of magnetization, magnetic susceptibility, and magnetostriction. An example of such application, in particular, is for the multistate multiferroic memories, since it is necessary to find a material that retains ME coefficient at zero bias.[17,41,42]

ACKNOWLEDGMENTS

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[1]L. D. Landau, E. M. Lifshitz, and L. P. Pitaevskii, *Electrodynamics of Continuous Media* (Pergamon, 1984).
[2]M. Fiebig, J. Phys. D: Appl. Phys. **38**, R123 (2005).
[3]W. Eerenstein, N. D. Mathur, and J. F. Scott, Nature **442**, 759 (2006).
[4]S. W. Cheong and M. Mostovoy, Nat. Mater. **6**, 13 (2007).
[5]D. Chiba, M. Sawicki, Y. Nishitani, Y. Nakatani, F. Matsukura, and H. Ohno, Nature **455**, 515 (2008).
[6]N. Hur, S. Park, P. A. Sharma, J. S. Ahn, S. Guha, and S. W. Cheong, Nature **429**, 392 (2004).
[7]K. Tahmasebi, A. Barzegar, J. Ding, T. S. Herng, A. Huang, and S. Shannigrahi, Mater. Des. **32**, 2370 (2011).
[8]G. Sreenivasulu, U. Laletin, V. M. Petrov, V. V. Petrov, and G. Srinivasan, Appl. Phys. Lett. **100**, 173506 (2012).
[9]C. Lu, P. Li, Y. Wen, A. Yang, C. Yang, D. Wang, W. He, and J. Zhang, J. Alloys Compd. **589**, 498 (2014).
[10]L. Chen, P. Li, Y. Wen, and Y. Zhu, J. Alloys Compd. **606**, 15 (2014).
[11]G. Q. Zhang, S. Dong, Z. B. Yan, Y. Y. Guo, Q. F. Zhang, S. Yunoki, E. Dagotto, and J. M. Liu, Phys. Rev. B **84**, 174413 (2011).
[12]T. Kimura, Annu. Rev. Condens. Matter Phys. **3**, 93 (2012).
[13]Y. Yamasaki, S. Miyasaka, Y. Kaneko, J. P. He, T. Arima, and Y. Tokura, Phys. Rev. Lett. **96**, 207204 (2006).
[14]G. Srinivasan, E. T. Rasmussen, B. J. Levin, and R. Hayes, Phys. Rev. B **65**, 134402 (2002).
[15]Y. Wang, J. Hu, Y. Lin, and C.-W. Nan, NPG Asia Mater. **2**, 61 (2010).
[16]M. H. Li, Z. G. Wang, Y. J. Wang, J. F. Li, and D. Viehland, Appl. Phys. Lett. **102**, 082404 (2013).
[17]S. K. Mandal, G. Sreenivasulu, V. M. Petrov, and G. Srinivasan, Phys. Rev. B **84**, 014432 (2011).
[18]Y. Tokura, S. Seki, and N. Nagaosa, Rep. Prog. Phys. **77**, 076501 (2014).
[19]M. Labusch, M. Etier, D. Lupascu, J. Schröder, and M.-A. Keip, Comput. Mech. **54**, 71 (2014).
[20]F. D. Ma, Y. M. Jin, Y. U. Wang, S. L. Kampe, and S. Dong, Acta Mater. **70**, 45 (2014).
[21]T. N. Yang, J.-M. Hu, C. W. Nan, and L. Q. Chen, Appl. Phys. Lett. **104**, 052904 (2014).
[22]C. L. Jia, T. L. Wei, C. J. Jiang, D. S. Xue, A. Sukhov, and J. Berakdar, Phys. Rev. B **90**, 054423 (2014).
[23]M. I. Bichurin and V. M. Petrov, Adv. Condens. Matter Phys. **2012**, 798310 (2012).
[24]A. J. Gualdi, F. L. Zabotto, D. Garcia, and A. J. A. d. Oliveira, J. Appl. Phys. **114**, 053913 (2013).
[25]F. L. Zabotto, A. J. Gualdi, J. A. Eiras, A. J. A. de Oliveira, and D. Garcia, Integr. Ferroelectr. **131**, 127 (2011).
[26]A. H. Morrish, *The Physical Principles of Magnetism* (Wiley, 2001).
[27]S. U. Blundell, *Magnetism in Condensed Matter* (OUP Oxford, 2001).
[28]M. U. Getzlaff, *Fundamentals of Magnetism* (Springer, 2007).
[29]J. v. Suchtelen, Philips Res. Rep. **27**, 10 (1972).
[30]C. W. Nan, M. I. Bichurin, S. X. Dong, D. Viehland, and G. Srinivasan, J. Appl. Phys. **103**, 031101 (2008).
[31]A. Ahlawat, S. Satapathy, V. G. Sathe, R. J. Choudhary, and P. K. Gupta, Appl. Phys. Lett. **103**, 252902 (2013).
[32]E. Granado, A. Garcia, J. A. Sanjurjo, C. Rettori, I. Torriani, F. Prado, R. D. Sanchez, A. Caneiro, and S. B. Oseroff, Phys. Rev. B **60**, 11879 (1999).
[33]E. W. Lee, Rep. Prog. Phys. **18**, 184 (1955).
[34]K. Cole and R. Cole, J. Chem. Phys. **9**, 341 (1941).
[35]B. Babić-Stojić, V. Jokanović, D. Milivojević, Z. Jagličić, D. Makovec, N. Jović, and M. Marinović-Cincović, J. Appl. Phys. **113**, 234311 (2013).
[36]K. Uchino, *Ferroelectric Devices* (Marcel Dekker, 2000).
[37]G. F. Dionne, *Magnetic Oxides* (Springer, 2010).
[38]J. C. Slonczewski, J. Appl. Phys. **32**, S253 (1961).
[39]J. C. Slonczewski, J. Phys. Chem. Solids **15**, 335 (1960).
[40]R. D. Greenough and E. W. Lee, J. Phys. D: Appl. Phys. **3**, 1595 (1970).
[41]J. F. Scott, Nat. Mater. **6**, 256 (2007).
[42]M. Bibes and A. Barthelemy, Nat. Mater. **7**, 425 (2008).