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Theory of the magnetoeletric effect in a lightly doped high-T_c cuprate

S. Mukherjee,[1,2,3] B. M. Andersen,[2] Z. Viskadourakis,[1] I. Radulov,[1] and C. Panagopoulos[1,4,5]

[1]*Institute of Electronic Structure and Laser, Foundation for Research and Technology Hellas, Heraklion 70013, Greece*

[2]*Niels Bohr Institute, University of Copenhagen, DK-2100 Copenhagen Ø, Denmark*

[3]*Niels Bohr International Academy, Niels Bohr Institute, University of Copenhagen, Blegdamsvej 17, DK-2100 Copenhagen Ø, Denmark*

[4]*Department of Physics, University of Crete, Heraklion 71003, Greece*

[5]*Division of Physics and Applied Physics, Nanyang Technological University, 637371, Singapore*

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In a recent study, Viskadourakis *et al.* (arXiv:1111.0050) discovered that extremely underdoped La_2CuO_{4+x} is a relaxor ferroelectric and a magnetoelectric material at low temperatures. It is further observed that the magnetoelectric response is anisotropic for different directions of electric polarization and applied magnetic field. By constructing an appropriate Landau theory, we show that a biquadratic magnetoelectric coupling can explain the experimentally observed polarization dependence on magnetic field. This coupling leads to several interesting low-temperature effects, including a feedback enhancement of the magnetization within the ferroelectric phase, and a predicted magnetocapacitive effect.

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The field of magnetoelectrics has witnessed intense theoretical and experimental progress in recent years, mainly driven by the discovery of the nonlinear magnetoelectric effect, i.e., the dominant magnetoelectric coupling is of higher order than a bilinear coupling between electric and magnetic fields.[1,2] Among the materials discovered with such physical properties are the so-called *birelaxors*.[3,4] These systems show both relaxor ferroelectric and relaxor magnetic properties and are associated with spin-charge coupling at a mesoscopic scale. Focusing on the parent high-T_c superconductor La_2CuO_{4+x} (LCO) with an exceptionally low carrier concentration $n = 10^{17}$ cm^{-3}, we have recently found that this material is in fact a ferroelectric at low temperatures.[5] More specifically, LCO has been shown to be a relaxor ferroelectric where the dielectric mode behavior is caused by freezing of randomly oriented polarized regions.[6,7] In addition, LCO exhibits a distinct magnetoelectric effect with a pronounced dependence of the polarization on an externally applied magnetic field. These experimental results about the nature of doped charge carriers in antiferromagnetic Mott insulators need to be theoretically explained. Here, we focus on the magnetoelectric effect discovered in Ref. 5, and show how nonlinear terms coupling polarization and magnetization naturally lead to the observed field effect.

Parent cuprate superconductors are two-dimensional (2D) antiferromagnets with weak interplanar exchange coupling giving rise to three-dimensional (3D) long-range Néel order.[8–10] In the case of LCO, the Cu spins are slightly canted out of the CuO_2 planes because of a finite Dzyaloshinskii-Moriya (DM) interaction existing in the low-temperature orthorhombic phase (LTO).[11] Though this allows a small ferromagnetic moment to build up on each CuO_2 plane, the net magnetic moment is zero since the moments in consecutive layers are oriented in opposite directions. On application of an external magnetic field a first-order spin-flop transition is observed at a critical magnetic field $H_{sf} \sim 5$ T.[11,12] Clear evidence for coupled spin and charge degrees of freedom in these systems comes from the observation of pronounced discontinuities in resistivity and dielectric constant at a magnetic field corresponding to H_{sf}.[5,13] Further evidence of such coupling is found in LCO by the possibility to detwin these crystals, e.g., by the application of an in-plane magnetic field.[14]

In the following we use a symmetry-based analysis to identify the particular magnetoelectric interaction terms that are responsible for spin-charge coupling in underdoped LCO. We further construct a Landau theory, and show how this model reproduces all the qualitative features of the magnetic field dependence of the polarization curves reported in Ref. 5.

Below approximately 530 K the crystal structure of LCO is LTO with space group *Cmca* (D_{2h}^{18}).[15] If $(\hat{\boldsymbol{x}},\hat{\boldsymbol{y}},\hat{\boldsymbol{z}})$ denote unit vectors along the crystal axes, then we define fractional translations by

$$\tau = \tfrac{1}{2}(a\hat{\boldsymbol{x}} + c\hat{\boldsymbol{z}})\tau' = \tfrac{1}{2}(a\hat{\boldsymbol{x}} + b\hat{\boldsymbol{y}}), \tag{1}$$

where a,b,c are the lattice constants. The symmetry elements of this crystal structure are then written as $G = G_0 + \tau G_0$, where G_0 contains the eight elements

$$E, I, \sigma_a, \sigma_b', \sigma_c', C_{2a}, C_{2b}', C_{2c}'. \tag{2}$$

Here E denotes the identity, I inversion about a Cu site, $\sigma_a,\sigma_b,\sigma_c$ reflections about the planes $x = a/2, y = b/2, z = c/2$, and C_{2a},C_{2b},C_{2c} are 180° rotations about the axes that emanate from the center of the unit cell. Primed elements must be complemented by translation τ' that is itself not a symmetry operation.

Taking into account the above-mentioned symmetry properties of the *Cmca* space group, the free energy can be expressed as a sum of three contributions

$$F = F_M + F_{MP} + F_P. \tag{3}$$

Here, F_M is the purely magnetic free energy, F_{MP} is the magnetoelectric contribution, and F_P is the polarization free energy. The magnetic free energy that accounts for the crystal structure of the LTO phase has been studied previously by,

e.g., Thio *et al.*,[16] and is given by

$$F_M = \frac{1}{2}\sum_{i=1}^{2}\left[\frac{\chi_{2D}^{-1}}{2}L_i^2 + \frac{1}{4}AL_i^4 + \frac{1}{6}BL_i^6 - CL_iM_i + \frac{\chi_0^{-1}}{2}M_i^2 - H_cM_i - H_{ab}L_i\right] + \frac{1}{2}J_\perp L_1L_2. \quad (4)$$

Here, the out-of-plane (c direction) [in-plane (a-b plane)] applied magnetic field is represented by H_c [H_{ab}]. The coefficients A, B, and C would in general be temperature dependent. The order parameter $M_i = (S_{Ai} + S_{Bi})/2$ is the ferromagnetic moment per spin with S_{Ai}, S_{Bi} being the sublattice spins in the ith plane, and $L_i = (S_{Ai} - S_{Bi})/2$ is the antiferromagnetic order parameter ($L_i||a$). The spins are slightly canted due to the DM interaction term $-CM_iL_i$, which causes them to lie in the a-c plane of the magnetic unit cell. The coupling between the different planes is included by the $J_\perp$ term.

The presence of an inversion symmetry in the space group of the crystal forbids any linear magnetoelectric effect[17] and the physics is dominated by nonlinear coupling terms. We can focus on the largest nonlinear terms by further noting that the experimentally observed polarization response is symmetric under inversion of the external magnetic field [i.e., $\boldsymbol{P}(\boldsymbol{H}) = \boldsymbol{P}(-\boldsymbol{H})$]. This implies that the dominant couplings are of even order in the magnetic order parameter. Hence, the following terms contribute to the magnetoelectric coupling:

$$F_{MP} = \sum_{\alpha,i}\left(\frac{\gamma_{1\alpha}}{2}L_i^2 + \frac{\gamma_{2\alpha}}{2}M_i^2 + \gamma_{3\alpha}M_iL_i\right)P_\alpha^2, \quad (5)$$

where the components for $\boldsymbol{P}$ run over $\alpha = (a,b,c)$ in the magnetic unit cell. The $\gamma_{1\alpha}$ and $\gamma_{2\alpha}$ terms have been introduced using symmetry arguments alone, but their microscopic origin can, e.g., originate from

$$H_{\rm int} = -\delta_{\rm me}\sum_{ij}\sum_{kl}\boldsymbol{S}_i\boldsymbol{S}_j\sigma_k\sigma_l. \quad (6)$$

This Hamiltonian describes a biquadratic coupling between spins $\boldsymbol{S}_{i,j}$ and structural pseudospins $\boldsymbol{\sigma}_{k,l}$.[18] A biquadratic coupling term has also been derived by Pirc *et al.*[19,20] In multiglass material such as doped $SrTiO_3$ (Ref. 21) or in $EuTiO_3$ (Ref. 22) these terms have been invoked to explain the observed magnetoelectric effect. Finally, the DM-induced biquadratic magnetoelectric coupling term with coefficient $\gamma_{3\alpha}$ has been used to explain magnetoelectricity in $BaMnF_4$.[23]

The polarization free energy is given by

$$F_P = \sum_\alpha\left(\frac{\chi_{e\alpha}^{-1}}{2}P_\alpha^2 + \frac{\beta}{4}P_\alpha^4\right) - \boldsymbol{E}\boldsymbol{P}. \quad (7)$$

Here, $\chi_{e\alpha}$ is the electric susceptibility for the α component of the polarization. Fourth-order terms have been included to obtain stable solutions, and $\boldsymbol{E}$ denotes the applied electric field. In its most general form, the free energy should also contain gradient terms since we are dealing with a relaxor system[20] as well as higher-order terms. However, since we are interested in the magnetoelectric effect close to the ferroelectric transition, it is reasonable to restrict our analysis to the above terms.

The solutions that determine $\boldsymbol{P}(\boldsymbol{H})$ are obtained by minimizing F with respect to the electric polarization and magnetic order parameters. In the case of LCO studied experimentally,

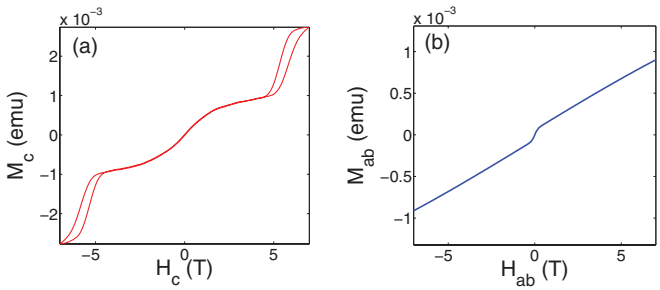

FIG. 1. (Color online) Experimental values of (a) out-of-plane magnetization and (b) in-plane magnetization at $T = 5$ K.

the Néel temperature is $T_N \sim 320$ K, which is much higher than the temperature at which the ferroelectric order sets in ($T_P \sim 4.5$ K).[5] Therefore, we evaluate F_M for the high-temperature phase with $\boldsymbol{P} = 0$, providing the following set of equations:

$$M_i = \chi_0(H_c + CL_i), \quad (8)$$

$$\chi_{2D}^{-1}L_1 + AL_1^3 + BL_1^5 + \tfrac{1}{2}J_\perp L_2 = CM_1 + H_{ab}, \quad (9)$$

$$\chi_{2D}^{-1}L_2 + AL_2^3 + BL_2^5 + \tfrac{1}{2}J_\perp L_1 = CM_2 + H_{ab}, \quad (10)$$

$$\left[\chi_{e\alpha}^{-1} + \sum_{i=1}^{2}\left(\gamma_{1\alpha}L_i^2 + \gamma_{2\alpha}M_i^2 + \gamma_{3\alpha}M_iL_i\right)\right]P_\alpha = -\beta P_\alpha^3. \quad (11)$$

The experimental magnetization curves at low temperatures ($\lesssim$30 K) have a glassy contribution that can be observed in Fig. 1(a) as a hump feature at low magnetic fields and a broadened spin-flop transition for out-of-plane magnetic fields H_c.[24] Such glassy behavior is absent in the magnetization response for in-plane applied magnetic fields H_{ab}, as seen from Fig. 1(b). These features cannot be obtained from the above equations, and hence to include them to the lowest order we simply take the experimental magnetization curves in Fig. 1 as input. This approximation, along with the temperature range at which we evaluate the magnetoelectric effect, leads to deviations from the parameters of the magnetic free energy from those used, e.g., by Thio *et al.*[16,25]

In terms of the following rescaled quantities $l_+ = \chi_0C(L_1 + L_2)/2$, $l_- = \chi_0C(L_1 - L_2)/2$, $M = (M_1 + M_2)/2$, $\gamma'_{1\alpha} = 2\gamma_{1\alpha}(\chi_0C)^{-2}$, $\gamma'_{2\alpha} = 2\gamma_{2\alpha}$, $\gamma'_{3\alpha} = 2\gamma_{3\alpha}(\chi_0C)^{-1}$, the polarization dependence on the applied magnetic field can be expressed as

$$\frac{P_\alpha(H_{ab})}{P_\alpha(0)} = \left\{1 + \frac{s_\alpha}{l_-^2(0)}[l_-^2(H_{ab}) - l_-^2(0) + l_+^2(H_{ab})]\right\}^{1/2}, \quad (12)$$

$$\frac{P_\alpha(H_c)}{P_\alpha(0)} = \left\{1 + \frac{s_\alpha}{l_-^2(0)}[l_-^2(H_c) - l_-^2(0) + g_\alpha M(H_c)^2 + (1 - g_\alpha - q_\alpha)l_+^2(H_c) + q_\alpha M(H_c)l_+(H_c)]\right\}^{1/2}, \quad (13)$$

where $s_\alpha = \lambda_\alpha l_-^2(0)/[\chi_{e\alpha}^{-1} + \lambda_\alpha l_-^2(0)]$, $g_\alpha = \gamma'_{2\alpha}/\lambda_\alpha$, and $q_\alpha = \gamma'_{3\alpha}/\lambda_\alpha$, with $\lambda_\alpha = \gamma'_{1\alpha} + \gamma'_{2\alpha} + \gamma'_{3\alpha}$. In general, all three

parameters s_α, g_α, and q_α will be temperature dependent. The temperature dependence of s_α primarily results from its relation to the electric susceptibility; hence an estimation of the amplitude of s_α can reveal the magnitude of anisotropy in the electric polarization.

In the case of an out-of-plane magnetic field H_c, the measured $P_c(H_c)$ increases with field and exhibits a pronounced hump at the spin-flop transition at $H_{\rm sf}$, as seen from Fig. 2(a). By contrast, $P_{ab}(H_c)$ decreases with increasing H_c but also exhibits a hump feature at $H_{\rm sf}$, as seen from Fig. 2(c). This behavior indicates a scenario where the coupling between the magnetic order and out-of-plane polarization P_c is attractive whereas the coupling with P_{ab} is repulsive. However, we find that the theoretical picture is more complex due to the presence of two competing magnetic orders $\boldsymbol{L}$ and $\boldsymbol{M}$ coupling to the electric polarization. For the out-of-plane magnetic field, we calculate the $P_\alpha(H_c)$ response using the magnetization data shown in Fig. 1(a). As seen from Figs. 2(b) and 2(d), we obtain qualitative agreement with both experimental polarization curves including the hump feature at $H_{\rm sf}$. Note that for the coupling to the magnetization we have set $g_a = g_b = g_c = -0.2$. Therefore, the source of anisotropy between $P_c(H_c)$ and $P_{ab}(H_c)$ is the DM-induced coupling term $q_\alpha P_\alpha^2 M_i L_i$ with $q_a = q_b = 0$ and $q_c = -6.58$.

In the case of an in-plane magnetic field H_{ab}, $P_\alpha(H_{ab})$ depends on only a single fitting parameter s_α that controls the magnitude of the polarization ratio, whereas the shapes of the theoretical curves are governed by the magnetic order parameter of the system. Further, we can deduce from Eq. (12) that the polarization primarily couples to the 2D antiferromagnetic order through a magnetoelectric interaction term $F_{MP} \sim \lambda_\alpha P_\alpha^2 L_i^2$. The anisotropy in polarization through the parameter λ_α for in-plane magnetic fields is primarily controlled by the DM-induced magnetoelectric coupling term $q_\alpha P_\alpha^2 M_i L_i$, since q_α undergoes the largest variation between in-plane and out-of-plane directions. The DM physics therefore plays an important role in generating an anisotropy between $P_c(\boldsymbol{H})$ and $P_{ab}(\boldsymbol{H})$ in La_2CuO_{4+x}.

Using the magnetization curve from Fig. 1(b), the solutions of $P_\alpha(H_{ab})$ are presented in Figs. 3(b) and 3(d). Though the theoretical curves have similar shapes, the scales are different in the two figures due to the difference in s_α values. Note that all theoretical curves have been plotted using magnetization data available at $T = 5$ K while the experimental curves correspond to $T = 2.5$ K, which accounts for the smaller scale of the theoretical curves. As seen from the plots, we find again qualitative agreement with the experimental data shown in Figs. 3(a) and 3(c), though the experimental plot in Fig. 3(a) has a steeper slope than theory. This steeper curve in the experimental curve could be the result of some history effect in these relaxor systems, a feature that has not been taken into account in our analysis. As in Fig. 2, the plots correspond to $s_c = 0.1$, $s_a = 0.074$, which leads to a small anisotropy in the electric susceptibilities $\chi_{e\alpha}^{-1}$ and therefore between the zero magnetic field values of in-plane polarization P_{ab} and the out-of-plane component P_c. This small anisotropy has been observed in experiments[5] and implies a much weaker anisotropy of the electric polarization compared to the magnetic order. This result lends support to a scenario where the nonstoichiometric oxygen dopants (charge carrier doping)

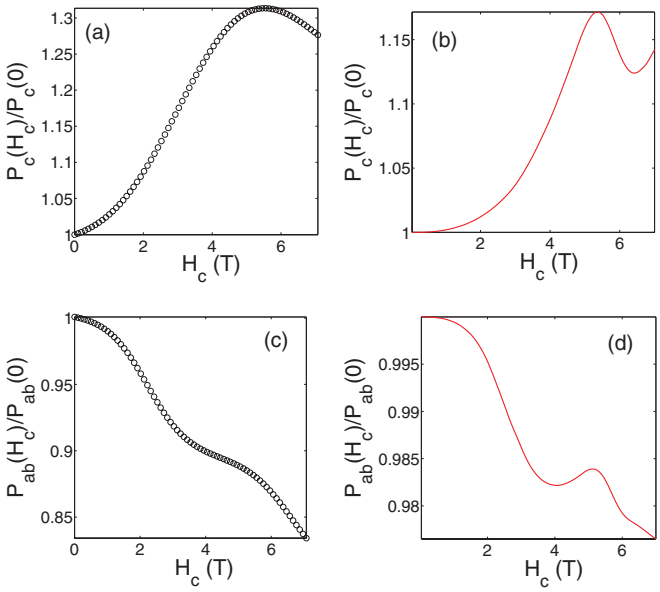

FIG. 2. (Color online) Polarization P_α vs out-of-plane magnetic field H_c. (a), (c) Experimentally measured values at $T = 2.5$ K. (b), (d) Theoretically calculated $P_\alpha(H_c)/P_\alpha(0)$ for $s_c = 0.1$, $s_a = 0.074$, $g_a = g_b = g_c = -0.2$, $q_c = -6.58$, and $q_a = q_b = 0$ using available experimental magnetization values at $T = 5$ K.

play an important role in generating the relaxor ferroelectricity in La_2CuO_{4+x}.[5]

We have observed experimentally that the magnetization shows a small enhancement below the temperatures where the ferroelectric order sets in.[26] This effect is in addition to the typical upturn in magnetization near the spin-glass freezing temperature.[27,28] We can study such a feedback effect of a finite polarization on the magnetization by minimizing F

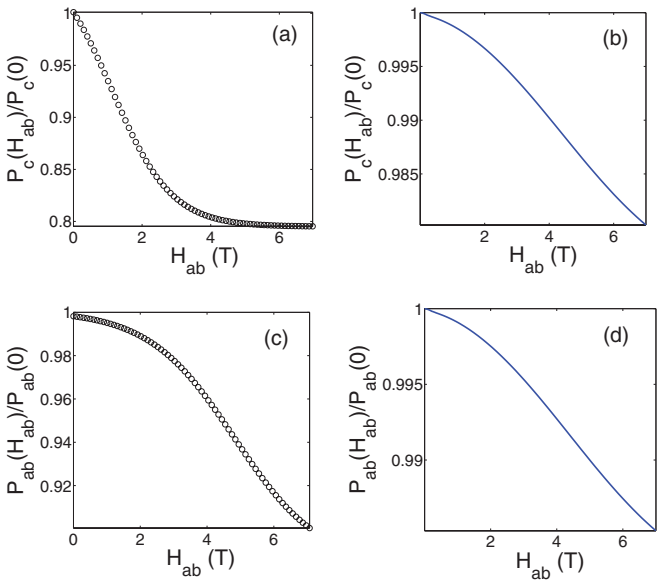

FIG. 3. (Color online) Polarization P_α vs in-plane magnetic field H_{ab}. (a), (c) Experimentally measured values at $T = 2.5$ K. (b), (d) Theoretically calculated $P_\alpha(H_{ab})/P_\alpha(0)$ for $s_c = 0.1$ and $s_a = 0.074$ using available experimental magnetization values at $T = 5$ K.

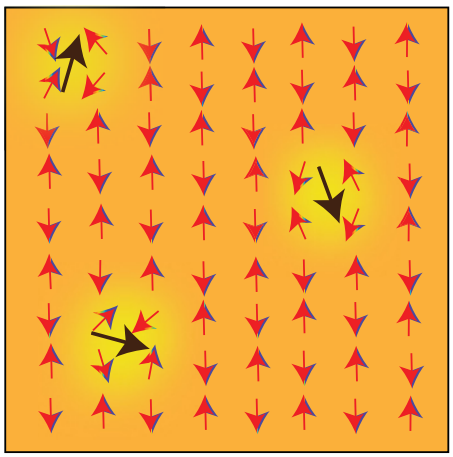

FIG. 4. (Color online) Real-space illustration of polar nanoregions with randomly oriented electric polarizations (large black arrows) at high T within the antiferromagnet (small red/gray arrows). Also shown are the distorted magnetic moments within a correlation length of the polar nanoregions (light yellow regions).

within the ferroelectric phase. This gives to lowest order

$$M_c = \frac{\chi_0 H_c + \left[1 - \chi_0 \sum_\alpha \gamma'_{3\alpha} P_\alpha^2(H_c)\right] l_+(H_c)}{1 + \chi_0 \sum_\alpha \gamma'_{2\alpha} P_\alpha^2(H_c)}, \quad (14)$$

$$M_{ab} = \frac{\left[1 - \chi_0 \sum_\alpha \gamma'_{3\alpha} P_\alpha^2(H_{ab})\right] l_+(H_{ab})}{1 + \chi_0 \sum_\alpha \gamma'_{2\alpha} P_\alpha^2(H_{ab})}. \quad (15)$$

Note that in this expression the relative sign of the coefficients can be determined from the relations $\gamma'_{3\alpha}/\gamma'_{2\alpha} = q_\alpha/g_\alpha > 0$. This also implies that since $\gamma'_{2\alpha} < 0$ in our model, it naturally causes an enhancement of magnetization due to the presence of a ferroelectric state. Additionally, we also find that this enhancement is present both in the in-plane and out-of-plane magnetization, with the relative size of the enhancement depending on the amplitude of polarization change with magnetic field.

A biquadratic coupling has been used to explain the observation of magnetocapacitive effects in materials such as doped $SrTiO_3$.[22] It is defined by the relation $\epsilon_\alpha = -\partial^2 F/\partial P_\alpha^2$ and hence requires at least quadratic terms in the polarization. For LCO the relative change in dielectric constant is given by

$$\Delta\epsilon_\alpha = [\epsilon_\alpha(\boldsymbol{H}) - \epsilon_\alpha(0)]/\epsilon_\alpha(0) = \Delta P_\alpha^2(\boldsymbol{H}). \quad (16)$$

A weak magnetocapacitive effect is therefore predicted in experiments at low temperatures. Note that in the above expression we would expect a small suppression in the permittivity for magnetic fields in the a-b plane.

The observation of relaxor ferroelectricity in underdoped LCO has been argued to originate from the formation of polar nanoregions (PNRs) around the nonstoichiometric oxygen dopants.[5] The formation of PNRs and the mechanism by which they condense into a ferroelectric phase is a well studied topic.[7,29,30] Though the relaxor physics in LCO naturally relates to the presence of dopants, the extremely low concentration of excess oxygen in the samples used in Ref. 5 may imply the presence of additional mechanisms for the PNRs to couple and undergo a spontaneous transition to long-range ferroelectric order. One may speculate that such mechanisms include subtle noncentrosymmetric structural distortions in the host lattice[12] and/or a tendency for the dopants to cluster and thereby reduce the inter-PNR distance. As shown in Fig. 4, in magnetic materials such as LCO the PNRs may also cause a distorted spin structure that could lead to a magnetoelectric effect through, e.g., geometric frustrations in the presence of a DM interaction and/or indirectly through coupling to strains. This physics has similarities to the observation of magnetoelectric behavior in a number of other relaxor ferroelectrics.[21,31]

In summary, we have shown that the magnetoelectric effect in extremely underdoped La_2CuO_{4+x} can be explained by biquadratic terms in the free energy. It is proposed that the microscopic origin of the ferroelectricity is caused by polar nanoregions generated by dopant ions. The discovery of ferroelectricity and a magnetoelectric effect in the cuprate materials due to charge carrier doping has sparked many questions for future studies. In particular, what happens at higher doping levels and what is the fate of the PNRs in the regime where the pseudogap and superconducting phases emerge?

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