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Large electrocaloric effect induced by the multi-domain to mono-domain transition in ferroelectrics

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The electrocaloric properties of multi-domain ferroelectrics are investigated using a phase field model. The simulation results show that the extrinsic contribution from the multi-domain to mono-domain transition driven by temperature significantly enhances the electrocaloric response. Due to the abrupt decrease of polarization in the direction of electric field during the domain transition, a large adiabatic temperature change is achieved for the ferroelectrics subjected to a tensile strain. Furthermore, the domain transition temperature can be tuned by external strains as the phase transition temperature. A compressive strain decreases the domain transition temperature while a tensile strain increases it. The large temperature change associated with the domain transition provides guidance to engineer domain structures by strain to optimize the electrocaloric properties of ferroelectric materials below the Curie temperature. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4873112>]

I. INTRODUCTION

The electrocaloric effect (ECE) refers to the temperature change of a material under adiabatic condition in response to an applied electric field, which is the converse pyroelectric effect that exists in all ferroelectric materials. The electrocaloric properties of ferroelectrics have been studied for several decades.^{1–3} In general, the ECE of bulk ferroelectric specimens is quite small, which causes a temperature change of merely 1–2 K. Since the observation of giant temperature change of 12 K in PZT thin film in 2006,⁴ the ECE of ferroelectrics have drawn considerable attention for their promising applications such as micro solid-state refrigeration with high efficiency and low cost.^{5–8}

The ECE of ferroelectrics is related to the temperature dependence of spontaneous polarization. Although the ECE occurs both above and below the Curie temperature,⁹ it becomes significant only near the phase transition where the polarization undergoes a dramatic change with temperature. The ECE induced by phase transition is the intrinsic electrocaloric response due to a temperature dependence of polarization magnitude within the domains. In addition to the intrinsic contribution, the ECE of ferroelectrics is also dependent on the extrinsic contribution due to the change of domain structures.^{10,11} Our previous phase field study revealed that there exists a multi-domain to mono-domain transition driven by temperature in strained ferroelectrics.¹² It is well known that a change of domain structure in ferroelectrics can significantly alter the piezoelectric and dielectric properties. Therefore, it is expected that the domain

transition has a significant influence on the electrocaloric response of ferroelectrics.

Direct characterization of ECE of ferroelectrics is often difficult to procure because of the small temperature changes produced by the large electric fields. The indirect method based on the thermodynamics theory becomes important to determine the ECE of ferroelectrics. In the early studies, most thermodynamic analysis on the ECE of bulk ferroelectrics and ferroelectric thin films were based on the mono-domain assumption.¹³ Using a mono-domain thermodynamic model, Akcay *et al.*^{14,15} calculated the magnitude of the ECE in BaTiO₃ thin films and investigated the influence of different electrical and mechanical boundary conditions, including the bias electrical field, lateral clamping, and misfit strain. The mono-domain thermodynamic model was also extended to study the effect of misfit strain on the ECE of “incipient” ferroelectric SrTiO₃ thin films,¹⁶ and to investigate the effect of grain size on the electrocaloric effect in dense BaTiO₃ nanocrystals.¹⁷ This kind of model provides a basis for estimating the intrinsic ECE of ferroelectrics, in which, however, the extrinsic contribution from domain transition is absent.

To model the ECE of multi-domain ferroelectrics, Shi *et al.*¹⁸ proposed a double-domain ferroelectric model by combining the Pauli’s master equation and the Landau-Devonshire theory, and successfully predicted the enhanced ECE above the Curie temperature in relaxor ferroelectrics.¹⁸ Based on the Landau-Devonshire theory, Karthik and Martin¹⁰ employed the field-dependent volume fraction of *c* domains in a ferroelectric thin film to include the extrinsic contribution of domain wall motion to the ECE, but the effect of inhomogeneous polarization at the domain wall was not considered. Recently, Li *et al.*^{19,20} investigated the ECE

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of multi-domain ferroelectrics by using phase field simulations that includes the effect of inhomogeneous polarization. Nevertheless, in their simulations, there is no domain transition driven by temperature. In this paper, we use a phase field model to investigate the influence of domain transition driven by temperature on the electrocaloric properties of multi-domain ferroelectrics subject to constant strains.

II. PHASE FIELD MODEL FOR FERROELECTRICS

Differing from the homogeneous assumption in mono-domain thermodynamic model, the thermodynamic energy in the present phase-field model is described in terms of spatially continuous but inhomogeneous polarization and strain, in which the domain wall energy is explicitly taken into account. The phase transition and domain transition are direct consequences of the minimization process of the total free energy of the entire system. The temporal evolution of spatially inhomogeneous polarization $\mathbf{P} = (P_1, P_2, P_3)$ can be obtained from the following time-dependent Ginzburg-Landau equation:^{21,22}

$$\frac{\partial P_i(\mathbf{r}, t)}{\partial t} = -L \frac{\delta F}{\delta P_i(\mathbf{r}, t)} \quad (i = 1, 2, 3), \quad (1)$$

where L is the kinetic coefficient, $F = \int_V f dv$ is the total free energy of the simulated system, and f is the total free energy density. The variation of $\frac{\delta F}{\delta P_i(\mathbf{r}, t)}$ at the right hand side of Eq. (1) represents the thermodynamic driving force for the spatial and temporal evolution of polarization, $\mathbf{r} = (x_1, x_2, x_3)$ denotes the spatial vector and t is time. The total free energy includes the bulk Landau energy, the domain wall energy, the elastic energy and the electric energy, which has the expression

$$f(P_i, P_{ij}, \varepsilon_{ij}, E_i) = f_L(P_i) + f_G(P_{ij}) + f_{ela}(P_i, \varepsilon_{ij}) + f_{ele}(P_i, E_i), \quad (2)$$

in which, ε_{ij} is the strain, E_i is the electric field, and P_{ij} denotes the spatial derivative of the i th component of the polarization vector, P_i , with respect to the j th coordinate and $i, j = 1, 2, 3$.

The bulk Landau energy density is usually described by a six-order polynomial of polarization as

$$\begin{aligned} f_L(P_i) = & \alpha_1(P_1^2 + P_2^2 + P_3^2) + \alpha_{11}(P_1^4 + P_2^4 + P_3^4) \\ & + \alpha_{12}(P_1^2 P_2^2 + P_2^2 P_3^2 + P_1^2 P_3^2) + \alpha_{111} \\ & \times (P_1^6 + P_2^6 + P_3^6) + \alpha_{112}(P_1^4(P_2^2 + P_3^2) \\ & + P_2^4(P_1^2 + P_3^2) + P_3^4(P_1^2 + P_2^2)) \\ & + \alpha_{123}(P_1^2 P_2^2 P_3^2), \end{aligned} \quad (3)$$

where α_{ij} and α_{ijk} are higher-order dielectric coefficients, and $\alpha_1 = (T - T_0)/2\varepsilon_0 \varepsilon c_0$ is the dielectric coefficient, in which T and T_0 are the temperature and Curie temperature, respectively, c_0 is the Curie constant and ε_0 is the dielectric constant of the vacuum. The domain wall energy is a function of polarization gradient. The lowest order of the gradient

energy density is used in the present work, which has the form as

$$\begin{aligned} f_G(P_{ij}) = & \frac{1}{2} G_{11}(P_{1,1}^2 + P_{2,2}^2 + P_{3,3}^2) \\ & + G_{12}(P_{1,1}P_{2,2} + P_{2,2}P_{3,3} + P_{1,1}P_{3,3}) \\ & + \frac{1}{2} G_{44}[(P_{1,2} + P_{2,1})^2 + (P_{2,3} + P_{3,2})^2 \\ & + (P_{1,3} + P_{3,1})^2] + \frac{1}{2} G'_{44}[(P_{1,2} - P_{2,1})^2 \\ & + (P_{2,3} - P_{3,2})^2 + (P_{1,3} - P_{3,1})^2], \end{aligned} \quad (4)$$

where G_{ij} and G'_{ij} are gradient energy coefficients. The gradient energy density gives the energy penalty for the spatially inhomogeneous polarization. The elastic energy density is calculated from

$$f_{ela}(P_i, \varepsilon_{ij}) = \frac{1}{2} c_{ijkl} \varepsilon_{ij}^{ela} \varepsilon_{kl}^{ela}, \quad (5)$$

where c_{ijkl} are the elastic modulus, the elastic strain has the form of $\varepsilon_{ij}^{ela} = \varepsilon_{ij} - \varepsilon_{ij}^0$, where ε_{ij} is the total strain and $\varepsilon_{ij}^0 = Q_{ijkl} P_k P_l$ is the stress-free strain or eigen strain induced by polarization, in which Q_{ijkl} is the electrostrictive coefficients. When there is an external strain $\varepsilon_{ij}^{(a)}$, it should be added into the elastic strain ε_{ij}^{ela} through superposition method. The electric energy includes the energy contribution from external electric field and the depolarization energy, which is expressed as

$$f_{ele} = -E_i P_i - \frac{1}{2} E_i^{dep} P_i, \quad (6)$$

where E_i is externally applied field, E_i^{dep} is the depolarization field, which is dependent on the polarization distribution in the simulated system.²¹ In the presence of an external electric field, the adiabatic temperature change is calculated by

$$\Delta T = -T \int_{E_a}^{E_b} \frac{1}{C_E(T, E)} \left(\frac{\partial P_i}{\partial T} \right)_E dE_i, \quad (7)$$

where $C_E(T, E)$ is the heat capacity per unit volume at constant electric field. Following Karthik and Martin,¹⁰ the contribution of the polarization to the heat capacity is neglected in the present work. The absolute value of the heat capacity of PbTiO₃ is approximately constant as 3.9×10^6 J/m³K in our calculation.

III. RESULTS AND DISCUSSION

A. Temperature-dependent domain transition

To investigate the influence of domain transition on the electrocaloric properties, a 2D PbTiO₃ ferroelectric model is employed in the present phase field simulation. The square ferroelectric is discretized by 32×32 grid points with a normalized cell size of $\Delta x_1^* = \Delta x_2^* = 1$. All the materials constants used in the simulation are the same as those of Ref. 12. For convenience, all the materials parameters are

normalized and the normalized equations are solved in the simulation. The normalized formula in the present study is the same as that used in the previous work.¹² The superscript * represents the normalized value in this paper. The periodic boundary condition is adopted in both the x_1 and x_2 directions. The stable domain structures are simulated in the ferroelectrics under different temperatures. The compressive strain $\varepsilon_{11}^{(a)} = -0.004$, $\varepsilon_{22}^{(a)} = 0$ and tensile strain $\varepsilon_{11}^{(a)} = 0.004$, $\varepsilon_{22}^{(a)} = 0$ are applied to the ferroelectrics, respectively. To obtain the stable polarization distribution at each temperature, the time step is set as $\Delta t^* = 0.04$ and the total number of steps is taken as 2000 in solving Eq. (1).

Figs. 1(a) and 1(b) show the temperature dependence of average polarization under different electric fields in the ferroelectrics with the inequiaxial compressive strain of $\varepsilon_{11}^{(a)} = -0.004$, $\varepsilon_{22}^{(a)} = 0$ and tensile strain of $\varepsilon_{11}^{(a)} = 0.004$, $\varepsilon_{22}^{(a)} = 0$, respectively. With such constant strain conditions, the polarizations form regular multi-domain structures with 90° domain walls in the absence of electric field according to our previous study.¹² In each curve of Fig. 1, there is a transition point where the first order derivative of polarization with respect to temperature is discontinuous. The points labeled by different letters, A_i and B_i ($i = 0, 1, 2$), correspond to the

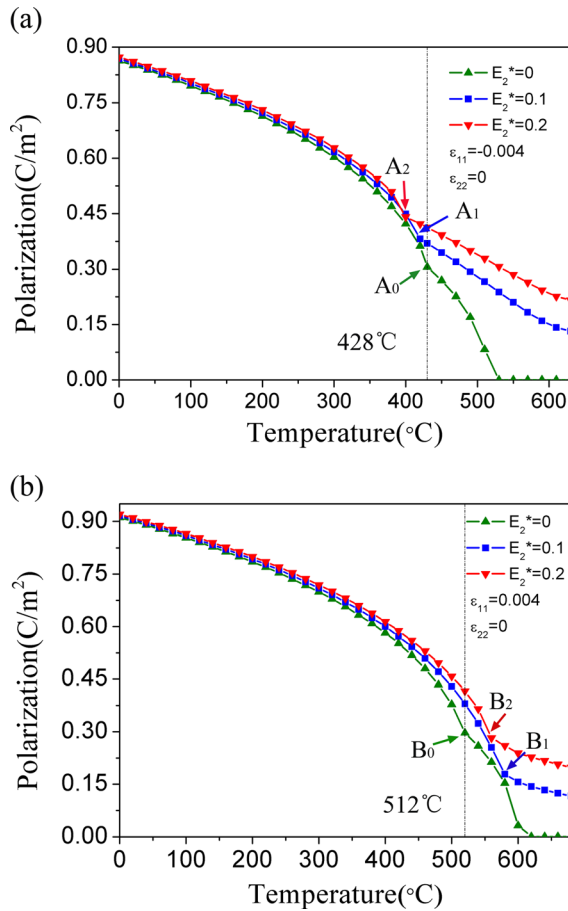


FIG. 1. Temperature dependence of the average polarization of the 2D PTO ferroelectrics subjected to different electric fields (a) with the compressive strain of $\varepsilon_{11}^{(a)} = -0.004$, $\varepsilon_{22}^{(a)} = 0$ and (b) with the tensile strain of $\varepsilon_{11}^{(a)} = 0.004$, $\varepsilon_{22}^{(a)} = 0$. The multi-domain to mono-domain transition points are indicated by the arrows. The normalized electric field $E_2^* = E_2/E_0$, in which $E_0 = 2.31 \times 10^8$ V/m is the characteristic electric field.

multi-domain to mono-domain transitions in the ferroelectrics. In the absence of external electric field, the temperatures for the transition from multi-domain to mono-domain are 428°C and 512°C for the compressive strain and tensile strain, respectively. When the normalized electric fields of $E_2^* = 0.1$ and $E_2^* = 0.2$ are applied, the domain transition temperature decreases for the compressive strain while it increases for the tensile strain, as shown by the transition points A_i and B_i in Fig. 1. When the electric field increases, the domain transition temperature decreases for both cases, indicating that the electric field has a significant influence on the domain transition temperature. Fig. 2 shows the detailed domain configurations before and after the transition points of A_0 and B_0 in Fig. 1 for the compressive strain and tensile strain, respectively. Before domain transition, the polarizations form a multi-domain pattern with 90° domain walls, which is shown in Figs. 2(a) and 2(c). The formation of 90° domain walls is to reduce the elastic energy because the simulated ferroelectric is under the constant strain condition. When the temperature increases, the multi-domain state becomes the mono-domain state at 428°C and 512°C for the ferroelectrics with the compressive strain and tensile strain, respectively, as shown in Figs. 2(b) and 2(d). The domain transition driven by temperature induces a dramatic change in the derivative of polarization with respect to temperature, which influences the adiabatic temperature change via Eq. (7).

B. Dependence of polarization on temperature

In order to understand the change of the derivative of polarization with respect to temperature, Figs. 3(a) and 3(b) give the variation of dP_2/dT along the diagonal line R_1 - R_2 of the square ferroelectrics in Fig. 2 under different temperatures and with the compressive strain and tensile strain,

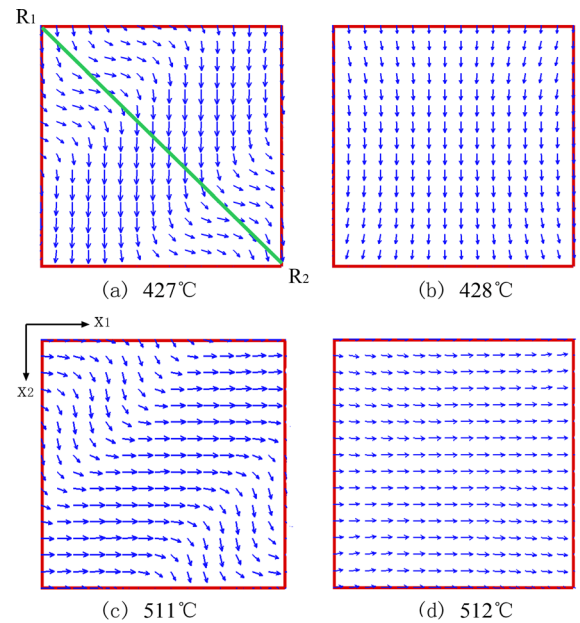


FIG. 2. The domain structures before (a), (c) and after (b), (d) the multi-domain to mono-domain transition under (a), (b) the compressive strain of $\varepsilon_{11}^{(a)} = -0.004$, $\varepsilon_{22}^{(a)} = 0$ and (c), (d) tensile strain of $\varepsilon_{11}^{(a)} = 0.004$, $\varepsilon_{22}^{(a)} = 0$ in the absence of electric field.

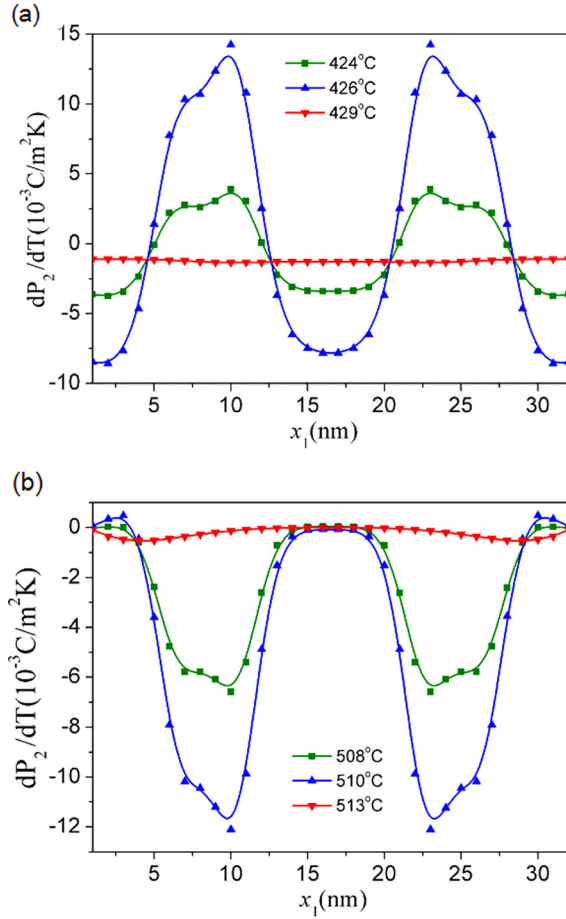


FIG. 3. The temperature dependences of dP_2/dT along the diagonal line R_1R_2 in Fig. 2 for the ferroelectric subjected to (a) the compressive strain of $\varepsilon_{11}^{(a)} = -0.004$, $\varepsilon_{22}^{(a)} = 0$ and (b) the tensile strain of $\varepsilon_{11}^{(a)} = 0.004$, $\varepsilon_{22}^{(a)} = 0$. The maximum value of dP_2/dT appears at the temperature of 426°C and 510°C , which are close to the corresponding domain transition temperatures.

respectively. It is found that dP_2/dT is highly inhomogeneous along the diagonal line of the square ferroelectrics with the multidomain states and under the temperature close to the domain transition temperature for both the compressive and tensile strains. However, the variation of dP_2/dT is quite small along the diagonal line in the mono-domain state when the temperature is larger than domain transition temperature, as shown by the curve with upwards triangles in Figs. 3(a) and 3(b). The maximum value of dP_2/dT appears at the temperature of 426°C and 510°C for the compressive and tensile strains, respectively, which are very close to their corresponding domain transition temperatures 428°C and 512°C . Interestingly, both positive and negative dP_2/dT exhibit a wide range of location for the compressive strain in Fig. 3(a). For the tensile strain of Fig. 3(b), however, the large value of dP_2/dT is negative in almost entire range. The negative dP_2/dT suggests that polarization component P_2 decreases with temperature, resulting in a positive temperature change according to Eq. (7). The results in Fig. 3 show that the compressive and tensile strains have different influence on the inhomogeneous distribution of the dP_2/dT . The inhomogeneous distribution of the dP_2/dT could induce an inhomogeneous distribution of adiabatic temperature change in the ferroelectrics. The advantage of the present phase field

model over the mono-domain thermodynamic method is that the spatially inhomogeneous adiabatic temperature change can be calculated, which gives more detailed information on the inhomogeneous electrocaloric properties.

C. Adiabatic temperature change

According to the definition of adiabatic temperature change of Eq. (7), the large dP_2/dT near the domain transition temperature results in a large adiabatic temperature change when the external electric field is applied. Figs. 4(a) and 4(b) show the adiabatic temperature change near the domain transition temperature under the electric field of $E_2^* = 0.1$ in the ferroelectrics subjected to the compressive and tensile strains, respectively. Due to the inhomogeneity of dP_2/dT , the adiabatic temperature change is spatially inhomogeneous along the diagonal line of the square ferroelectrics. For comparison, the adiabatic temperature change calculated from the mono-domain model is also given. The huge adiabatic temperature change with the maximum value of $\Delta T = 10\text{ K}$ exhibits at 510°C for the tensile strains, respectively, which is much larger than the value of 1.85 K predicted by mono-domain model, and is close to the giant ECE of $\Delta T = 12\text{ K}$ that has been experimentally observed in $\text{PbZr}_{0.95}\text{Ti}_{0.05}\text{O}_3$ thin films.⁴ The locations of maximum ECE

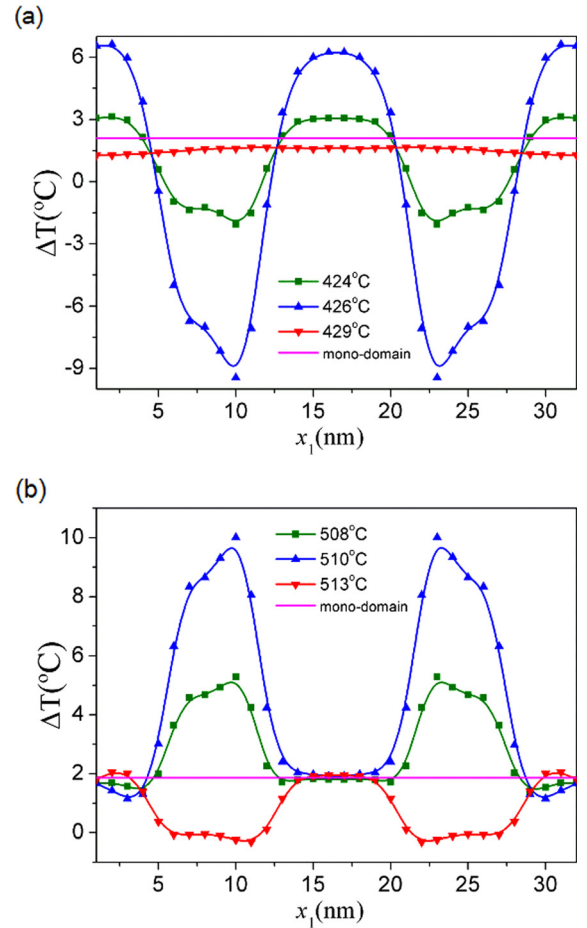


FIG. 4. The adiabatic temperature change near the domain transition temperature along the diagonal line R_1R_2 in Fig. 2 under the electric field of $E_2^* = 0.1$ in the ferroelectrics subjected to (a) the compressive strain of $\varepsilon_{11}^{(a)} = -0.004$, $\varepsilon_{22}^{(a)} = 0$ and (b) the tensile strain of $\varepsilon_{11}^{(a)} = 0.004$, $\varepsilon_{22}^{(a)} = 0$.

coincide with those of maximum dP_2/dT in Fig. 3, indicating that the large polarization change with temperature induced by domain transition makes a major contribution to the large ECE. Due to the influence of dP_2/dT , the ECE becomes smaller when the temperature deviates from the domain transition temperature, as shown by the curve of $T = 424^\circ\text{C}$ in Fig. 4. When the temperature is higher than the domain transition temperature, the ECEs of the mono-domain state at 429°C and 513°C for compressive and tensile strain, respectively, are much smaller than those of multi-domain state. These results imply that the domain transition is responsible for the appearance of huge ECE of the ferroelectric only in a specific range of temperature. The mechanism of the huge ECE induced by domain transition is similar to that the phase transition induced large ECE in ferroelectrics. In particular, the domain transition can be regarded as a generalized phase transition, it is reasonable that the domain transition has the similar effect on the ECE of ferroelectrics as the phase transition. Furthermore, the domain transition temperature is often lower than the phase transition temperature, which enlarges the temperature range where the large ECE exhibits. Similar to the phase transition temperature, the domain transition temperature with large ECE can be tuned by external strains. Fig. 4(b) shows that the maximum adiabatic

temperature change is located at 510°C for the ferroelectric subjected to a tensile strain, which is much smaller than the temperature of 426°C for the compressive strain. In addition, the average adiabatic temperature change for the tensile strain is 4.5 K while it is close to zero for the compressive strain. The results imply that a tensile strain perpendicular to the electric field increases the adiabatic temperature change for multi-domain ferroelectrics. In practice, the measured ECE is the average temperature change of the entire sample. To examine the average ECE of the whole simulated system, the average temperature changes as a function of applied electric field are given in Fig. 5. For the tensile strain, the largest average temperature change of the entire model is 16.7 K, which is located at 510°C near the domain transition temperature as shown in Fig. 5(b). On the other hand, a negative average temperature change of -1.8 K is found at 425°C near the domain transition temperature for the compressive strain, as shown in Fig. 5(a). The calculated small negative average temperature change under the compressive strain is consistent with the local temperature change in Fig. 4(a), in which both the negative and positive temperature changes occurred and the negative one has a larger magnitude than the positive one. The second peak in the curve of Fig. 5(a) is induced by the ferroelectric phase transition. The large average temperature change of the entire model in Fig. 5(b) is also consistent with the result of Fig. 4(b), which confirms that the domain transition can induce large ECE in multi-domain ferroelectrics subjected to a mechanical strain.

IV. CONCLUSIONS

In summary, the large ECE induced by the multi-domain to mono-domain transition in ferroelectrics is investigated by using a phase field model. The huge ECE induced by domain transition exhibits only in a range of temperature close to the domain transition temperature, which is similar to that the phase transition induced large ECE in ferroelectrics. The domain transition temperature with large ECE can be tuned by external strains as the phase transition temperature. The domain transition temperature is lower than the Curie temperature, which enlarges the range of temperature where the large ECE exhibits in ferroelectrics. A tensile strain not only increases the domain transition temperature but also induces a much larger average temperature change than a compressive one. The large temperature change associated with the domain transition provides another way to design the electrocaloric properties of ferroelectric materials through domain engineering.

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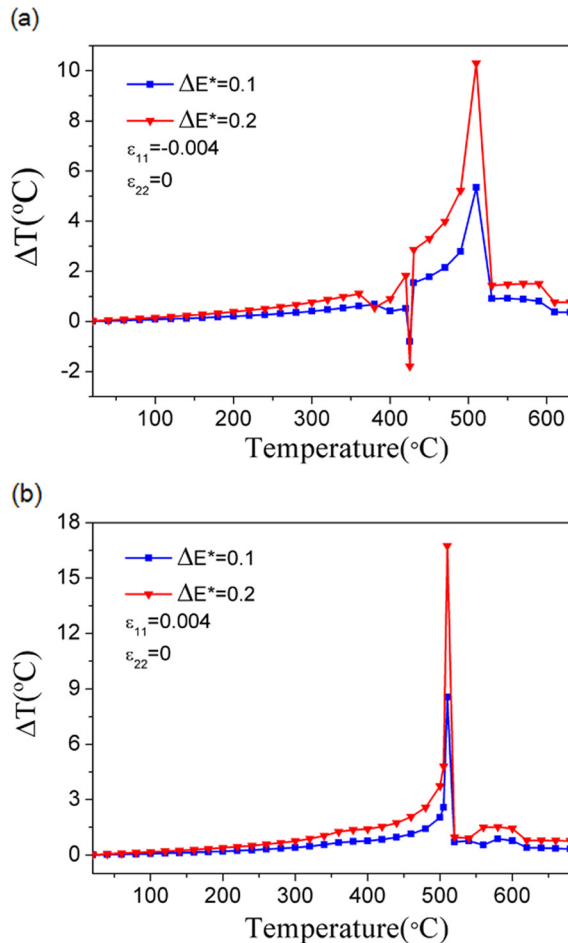


FIG. 5. Temperature dependence of the average temperature change in the ferroelectrics subjected to (a) the compressive strain of $\varepsilon_{11}^{(a)} = -0.004$, $\varepsilon_{22}^{(a)} = 0$ and (b) the tensile strain of $\varepsilon_{11}^{(a)} = 0.004$, $\varepsilon_{22}^{(a)} = 0$.

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