Ferroelastic Switching in PZT Thin Films: Phase-field Modeling

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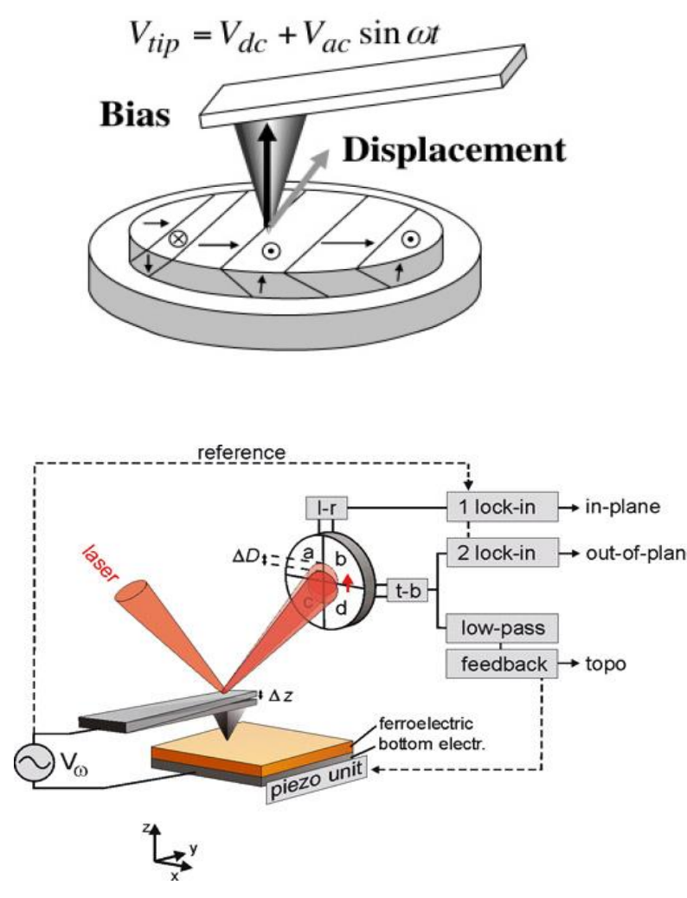
Background

Piezoresponse force microscopy (PFM) [1]



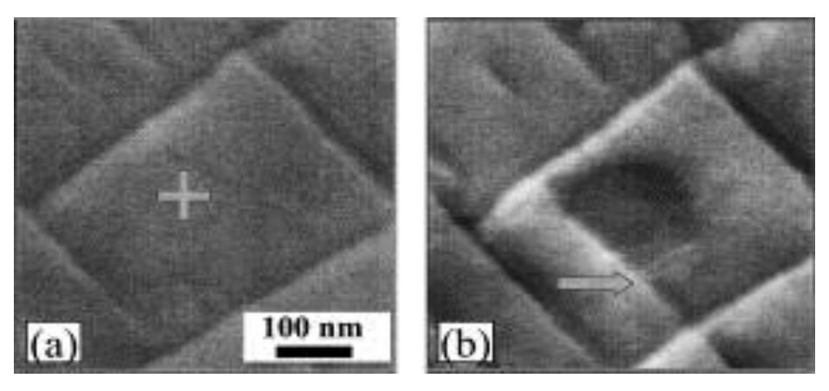
- ❖ Basic principle: Converse piezoelectric effect

$$X_i = d_{ki}E_k$$

- ❖ Applications: domain Imaging, domain patterning, studies of domain dynamics, spectroscopy and switching spectroscopy mapping
- ❖ Tip geometry: Lorentz distribution of potential

$$\varphi(x,y) = \varphi_0 \frac{\gamma^2}{\gamma^2 + (x-x_0)^2 + (y-y_0)^2}$$

PFM tip induced 90° switching [2]



- ❖ Local 90° switching has been observed in experiments
- ❖ Lack of detailed explanation and theoretical simulations

Methodology

❖ Time-dependent Ginzburg-Landau equations [3]

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

$$F = \int (f_{bulk} + f_{grad} + f_{elas} + f_{elec})dV$$

$$f_{bulk} = \frac{1}{2}\alpha_{ij}P_iP_j + \frac{1}{4}\gamma_{ijkl}P_iP_jP_kP_l + \frac{1}{6}\omega_{ijklmn}P_iP_jP_kP_lP_mP_n + \ldots$$

$$f_{grad} = \frac{1}{2}G_{ijkl}P_{i,j}P_{k,l}$$

$$f_{elas} = \frac{1}{2}c_{ijkl}(\varepsilon_{ij} - \varepsilon_{ij}{}^0)(\varepsilon_{kl} - \varepsilon_{kl}{}^0)$$

$$f_{elec} = -\frac{1}{2}\varepsilon_0\kappa_{ij}E_iE_j - E_iP_i$$

❖ Semi-implicit Fourier scheme [4]

$$\overline{P_i^*}^{n+1}(\vec{k},t) = \frac{\overline{P_i^*}^n(\vec{k},t) + \Delta t\,\overline{f_i^*}(\vec{k},t)}{1+\Delta t k_{Gi}^2}$$

❖ Boundary conditions

$$\varphi_{bottom} = 0, \varphi_{top} = \varphi(x,y)$$

$$\sigma_{i3}|_{x=h_f} = 0$$

$$u_i|_{substratebottom} = 0$$

❖ Additional conditions

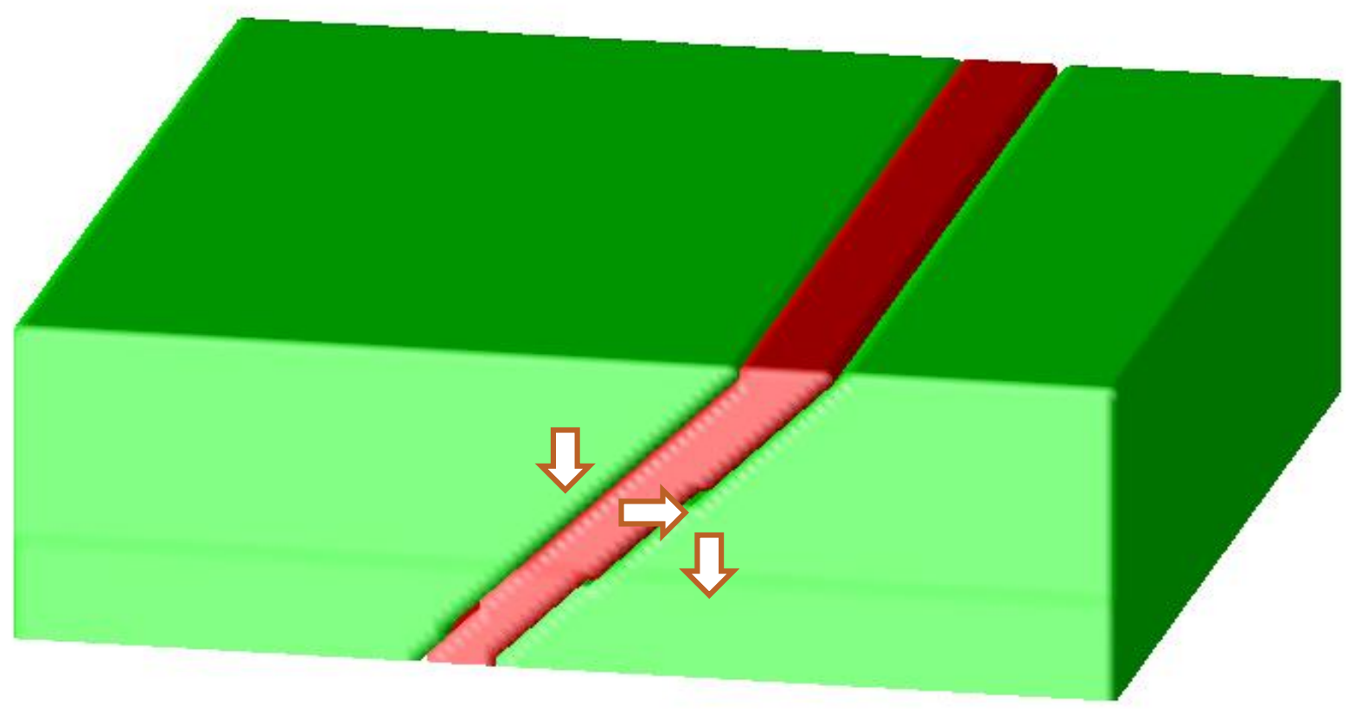
$$\varepsilon_0\kappa_{ij}\frac{\partial^2\varphi}{\partial x_i\partial x_j} = \frac{\partial P_i}{\partial x_i}$$

$$\sigma_{ij,j} = 0$$

$$\sigma_{ij} = c_{ijkl}(\varepsilon_{ij} - \varepsilon_{ij}{}^0)$$

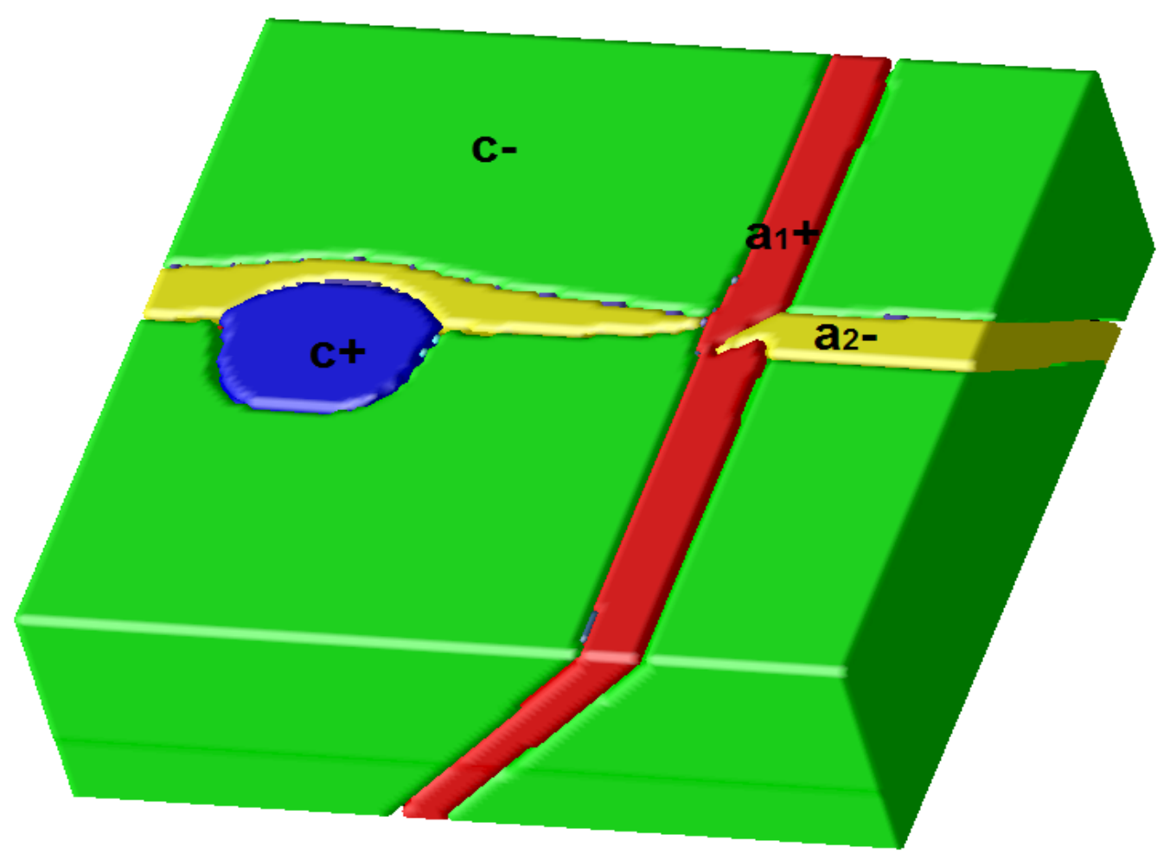
Ferroelastic switching and domain wall influence

(1) Initial domain configuration



Apply -2.2 V

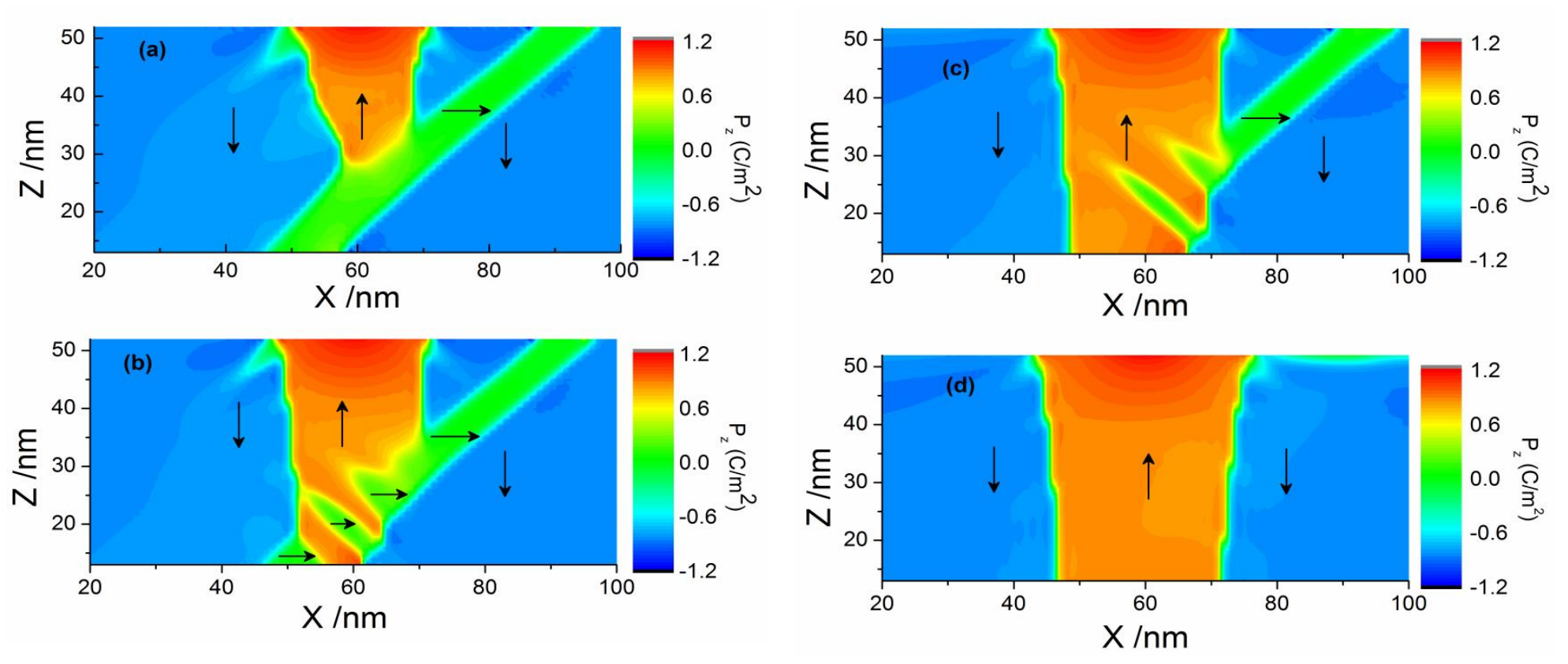
(2) 90° switching far from domain wall

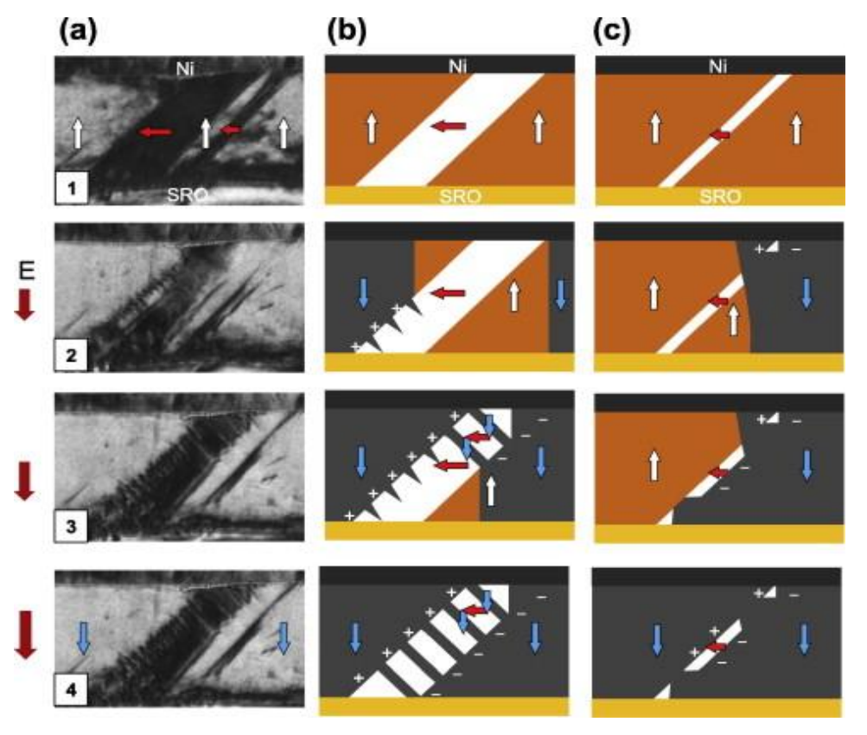


- ❖ We stabilize the *a*/*c* configuration as our initial domain
- ❖ The *a* domain tilts 45 degree with the *c* domain

- ❖ When switched at high bias, 90° switching is observed along with local 180° switching

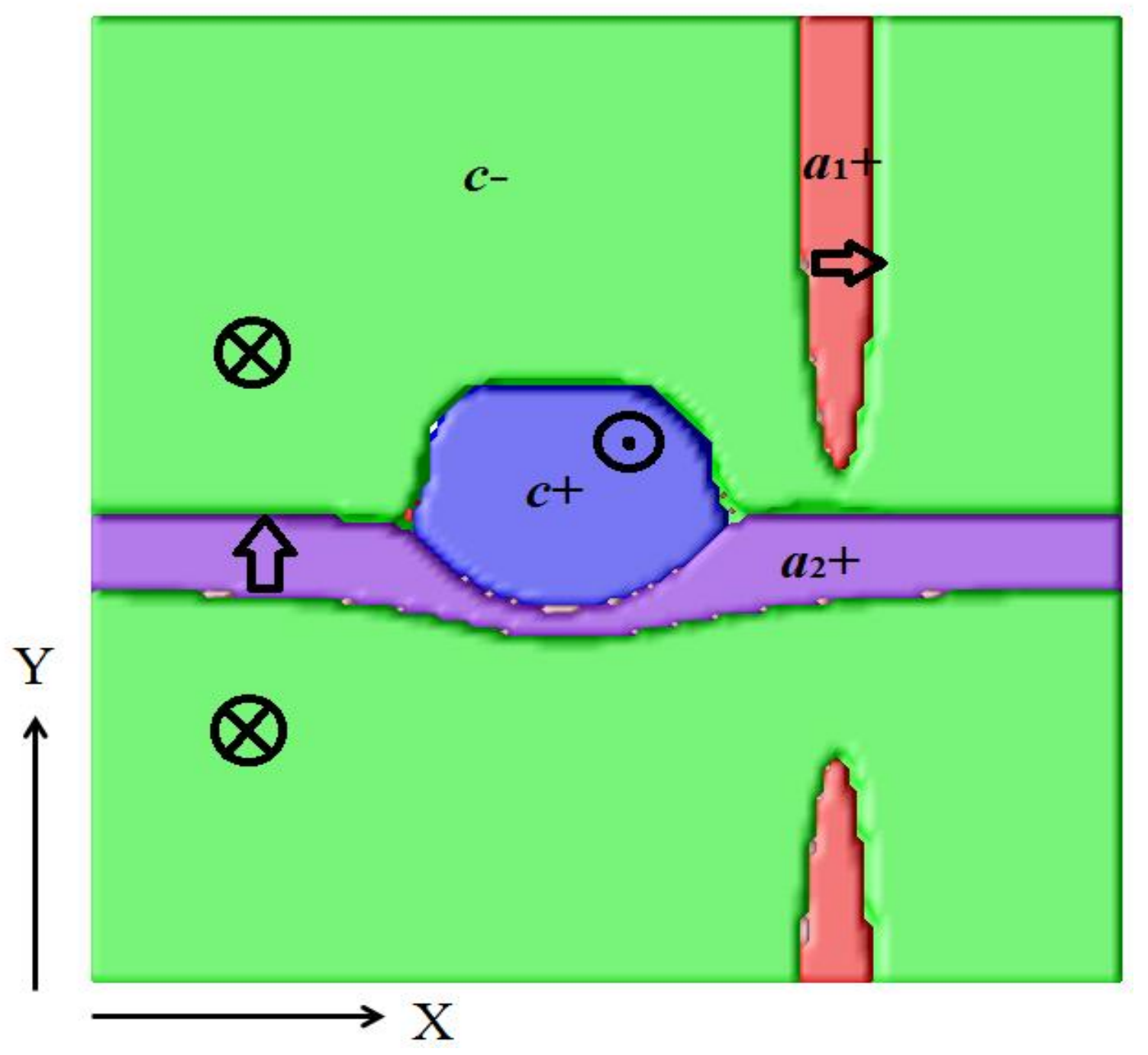
(3) 90° domain wall influence





- ❖ *c*+ domain grows and interacts with the a_1+ domain, forming tail to tail configuration with charged domain wall.
- ❖ Part of the a_1+ domain switches to *c*+ domains, remaining a_1+ domains form triangularly shaped stripe *a*-domains to minimize the bound charges
- ❖ Growth of the switched *c* domain occurs through thinning and elongation of triangularly shaped stripe *a*-domains
- ❖ Similar switching and growth process has been observed in experiments [5]

(4) Final domain structure with tip near domain wall



References

(1) S. V. Kalinin, A. N. Morozovska, L. Q. Chen, and B. J. Rodriguez, *Rep. Prog. Phys.* **73** (5), 056502 (2010).
(2) L. Chen, J. Ouyang, C. S. Ganpule, et al., *Appl. Phys. Lett.* **84** (2), 254 (2004).
(3) Y. L. Li, S. Y. Hu, Z. K. Liu, et al. *Acta Mater.* **50** (2), 395 (2002).
(4) L. Q. Chen and J. Shen, *Comput. Phys. Commun.*, **108** (2-3), 147 (1998).
(5) J. K. Lee, G. Y. Shin, K. Song, et al. *Acta Mater.* **61** (18), 6765 (2013).
(6) Z. J. Hong, J. Britson, J. –M. Hu and L. –Q. Chen, *Acta Mater.* **73** (2014): 75-82

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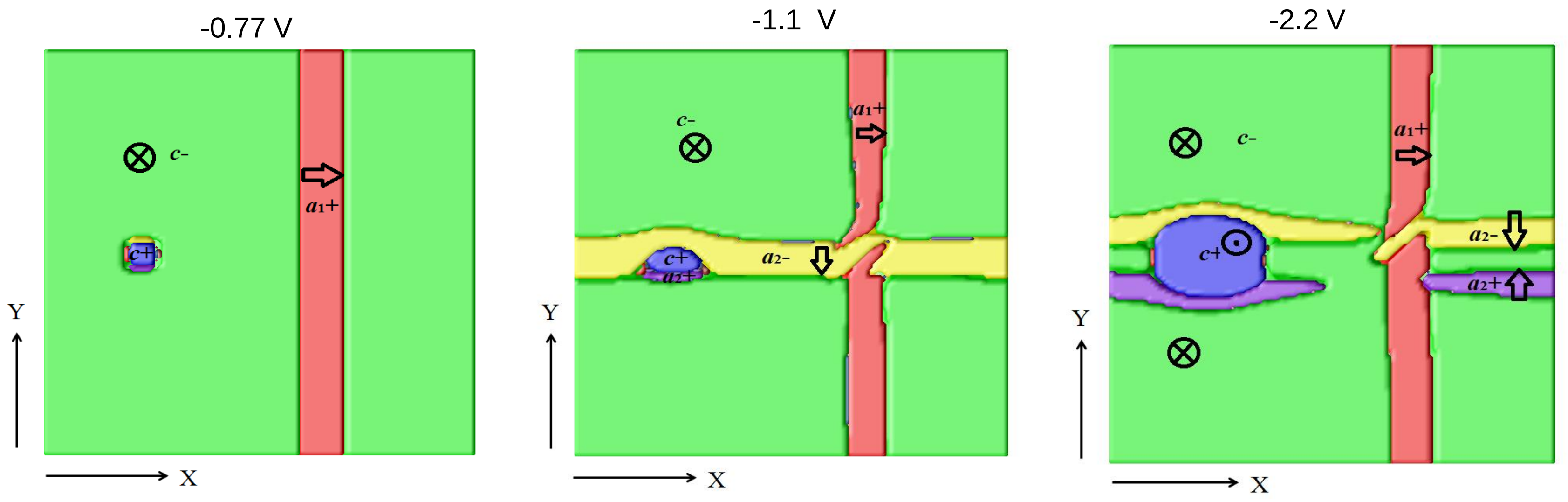
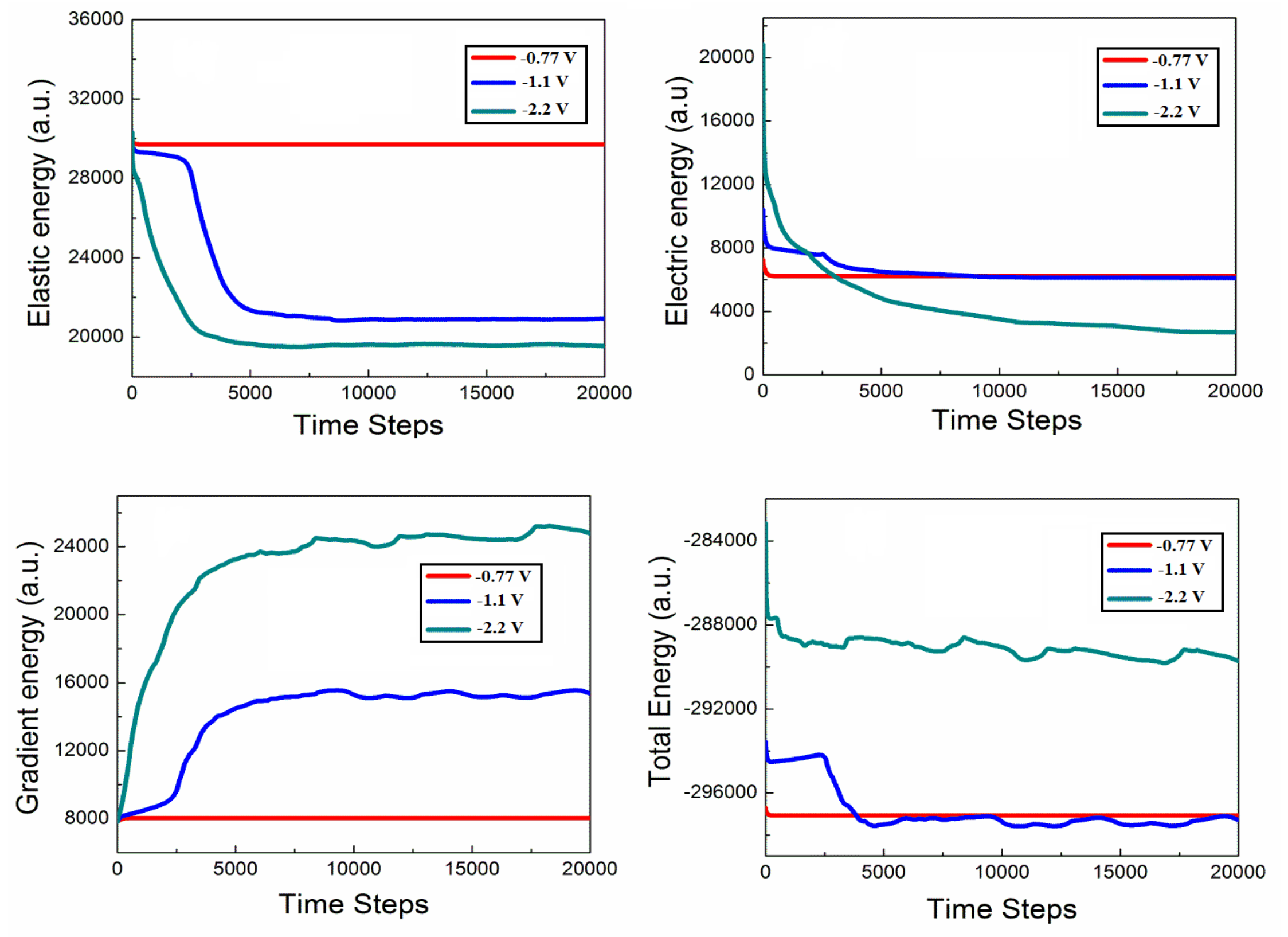
Switching under different strain and applied field

(1) Coherency strain -0.4%



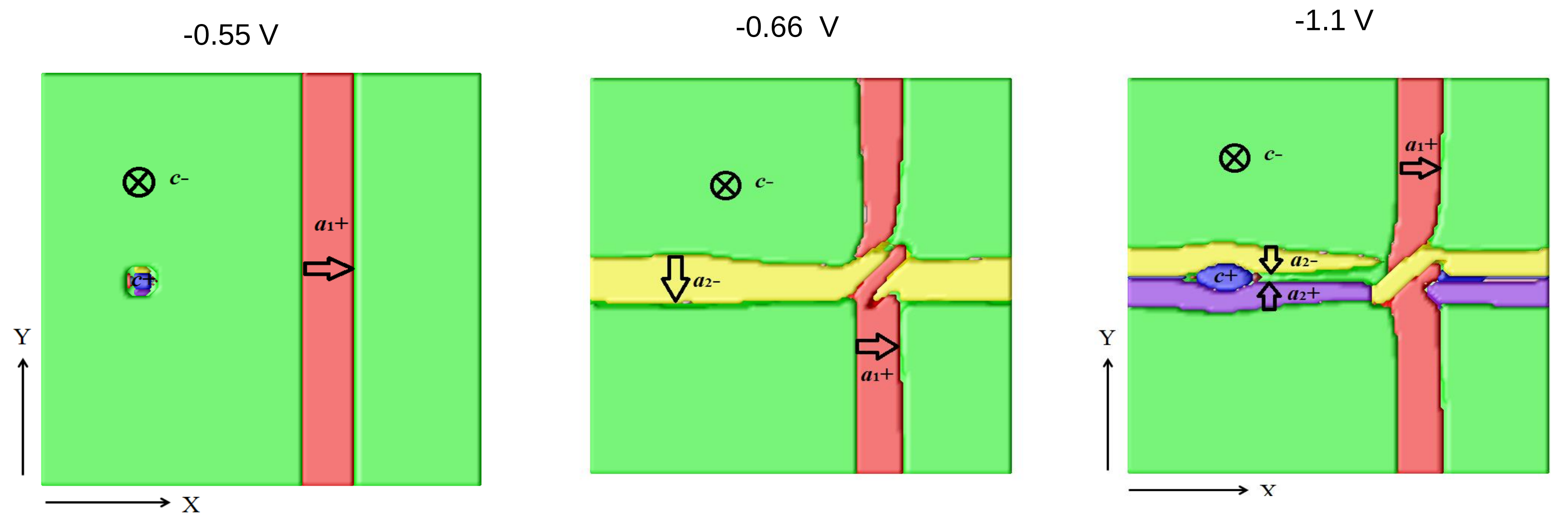
- ❖ 3 different switching patterns are observed:
  - ❖ Growth of *c* domains
  - ❖ Growth of *c* domain and one *a* domain
  - ❖ Growth of *c* domain and two *a* domains

- ❖ Dominated by 180 degree switching
- ❖ Dominated by 90 degree switching
- ❖ Dominated by 180 degree switching again



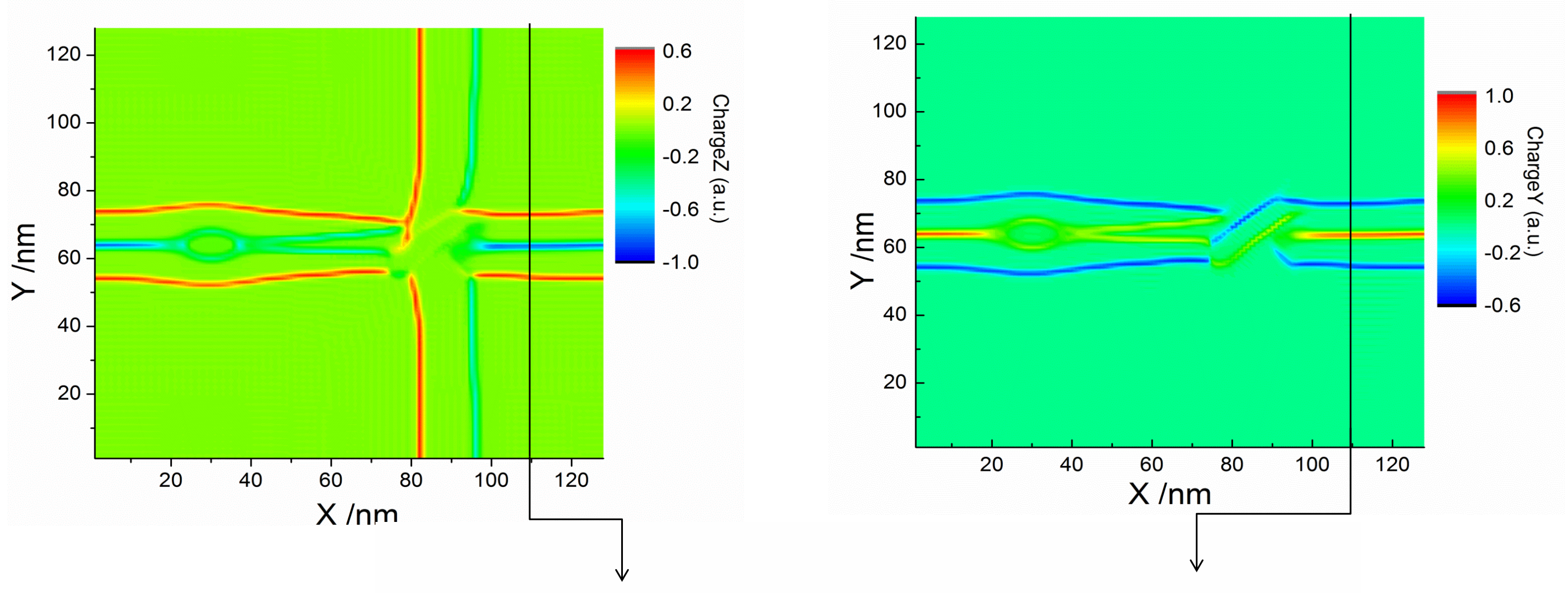
- ❖ With increasing bias, elastic energy drops with the growth of *a* domains
- ❖ Reductions in the elastic energy become smaller for the growth of additional *a*-domains
- ❖ The gradient energy increases dramatically as more and more domain walls are involved

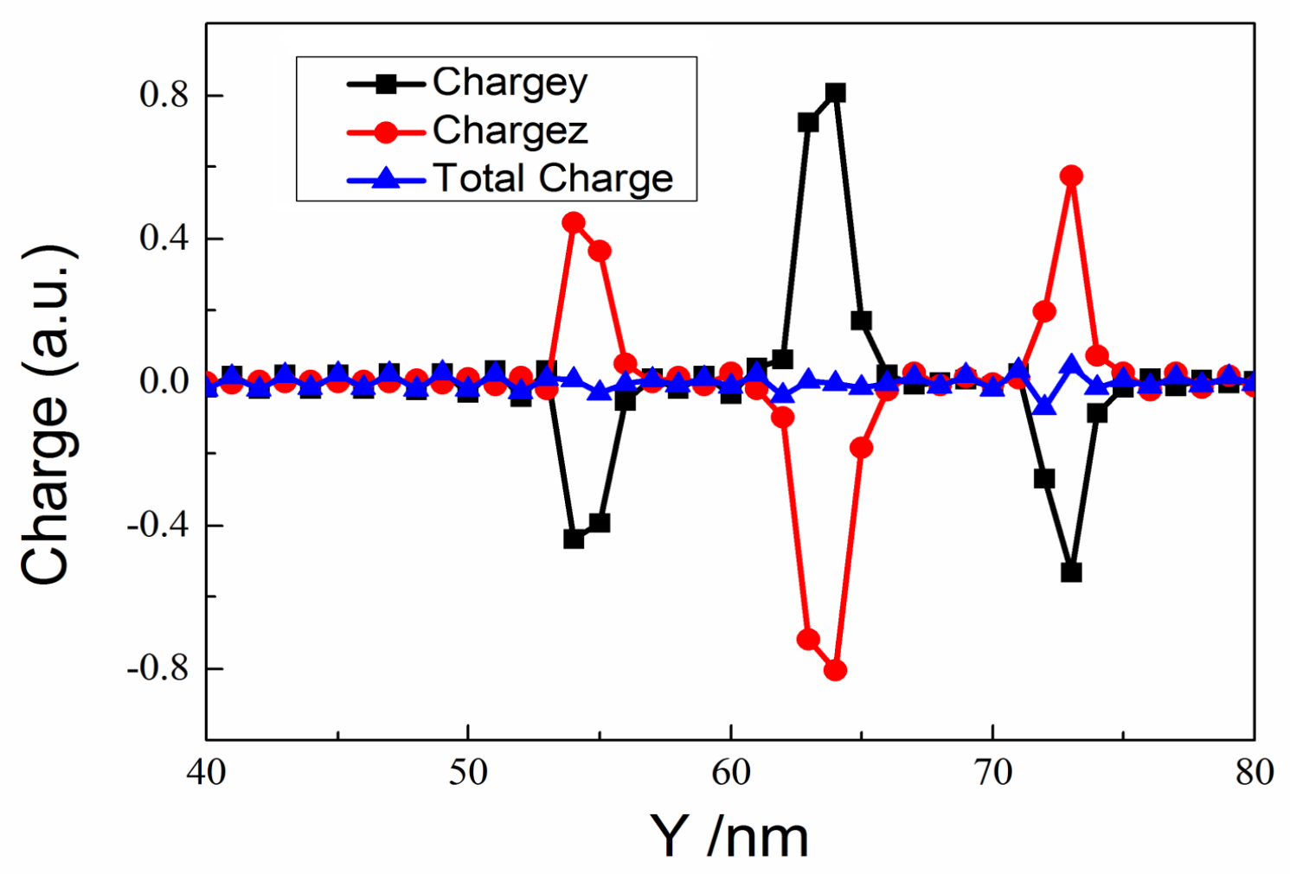
(2) Coherency strain -0.3%



- ❖ The bias for 90 degree switching is decreased compare to -0.4% case
- ❖ When we increase the bias further, 90 degree switching from *c*- to *a*- is favored
- ❖ At intermediate bias, both 90 degree and 180 degree switching are favored
- ❖ 180 degree switching occur between the *a* domains

(3) Charge distribution with applying bias -1.1 V





- ❖ As the two a_2-domains get close, the high P_2 gradient induces high charge
- ❖ This charge is compensated by chargez, which means the charge induced by P_3 gradient
- ❖ Thus 180° switching is favored between the two a_2-domains due to charge compensation.

Conclusions

- ❖ 90° switching is favored during the local 180° switching
- ❖ Decreasing the magnitude of compressive strain could lead to an increase in elastic energy, which favors the growth of *a*-domains
- ❖ At relatively high bias, however, 180° switching is favored due to a rapid decrease in electrostatic energy.
- ❖ At medium bias, with low magnitude of compressive strain, due to charge compensation, 180° switching may occur far away from PFM tip near the two newly-grown *a*-domains.