Reversible multi- to single-domain transition in $Ba(Zr_{0.2}Ti_{0.8})O_3$-x$(Ba_{0.7}Ca_{0.3})TiO_3$ ferroelectrics under poling conditions

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Outline

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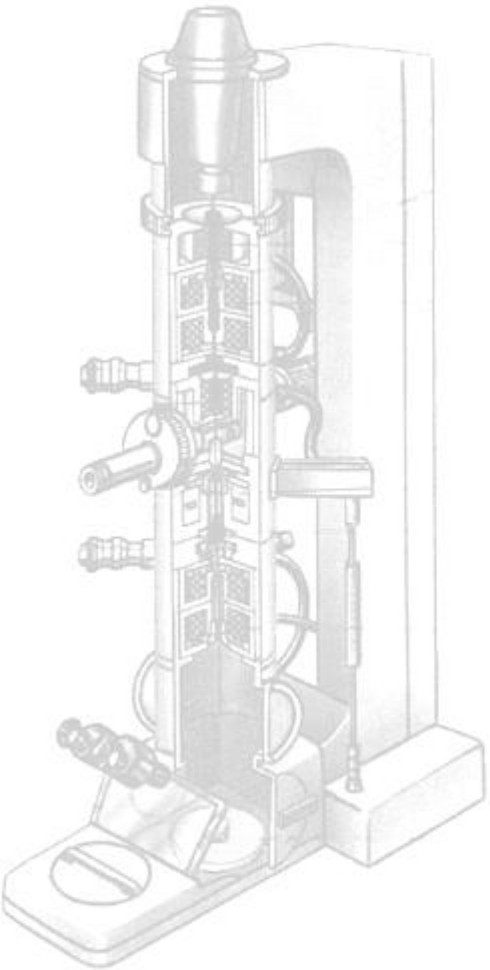
$Ba(Zr_{0.2}Ti_{0.8})O_3$-x$(Ba_{0.7}Ca_{0.3})TiO_3$



- Basics: Structure & phase diagram
- Materials and methods
- TEM Results on BZT-xBCT:
  - → *In-situ* electric field investigations
  - → *In-situ* electric field + cold stage investigations
- Summary

Perovskite structure

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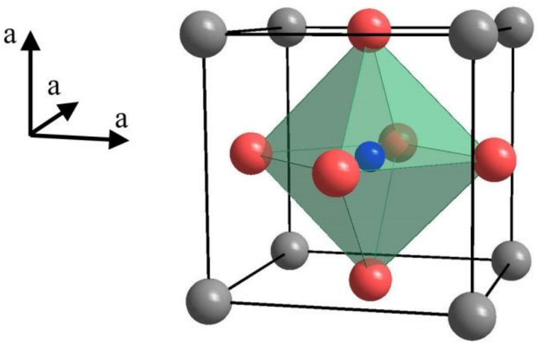
Perovskite has a cation on the A-site and a different cation on the B-site, which is octahedrally coordinated by anions.

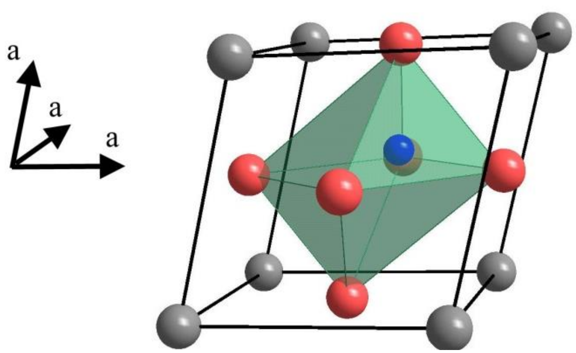
Pm$\bar{3}$m



Ba^{2+}/Ca^{2+}
Ti^{4+}/ Zr^{4+}
O^{2-}

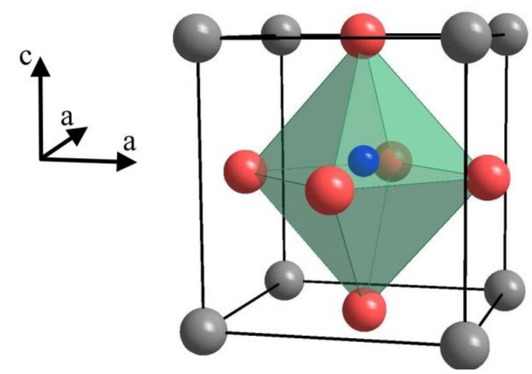
a = 4,008 Å
α, β, γ = 90°

R3m



a = 4,007 Å
α, β, γ = 89,95°

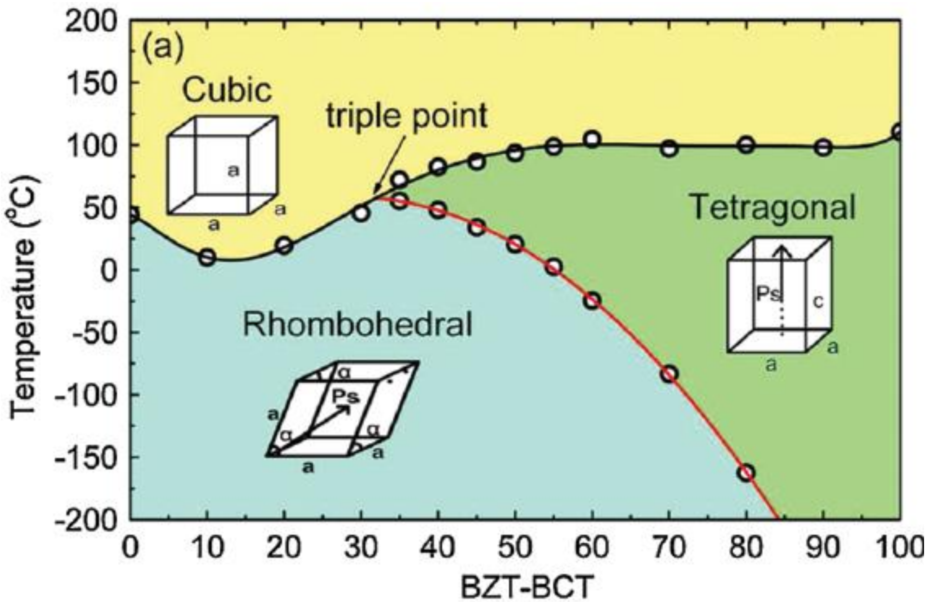
P4mm



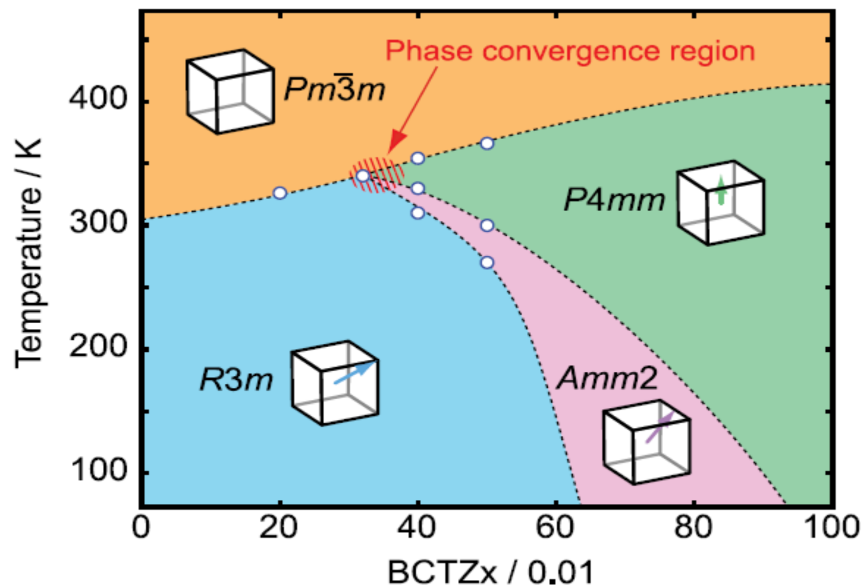
a = 4,005 Å, c = 4,011 Å
α, β, γ = 90°

Phase diagrams

Liu et al. PRL 103 (2009)



Keeble et al. APL (2013)



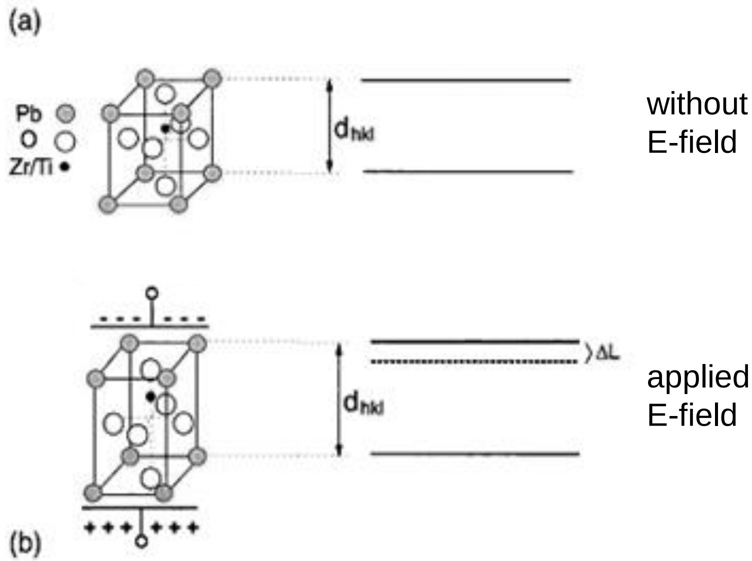
The phase diagram is characterized by a MPB separating a ferroelectric R and T phases. The most important feature of the BZT-BCT system is the existence of a C-R-T triple point.

The observed orthorhombic Amm2 phase persists all the way to the phase convergence region.

Intrinsic and extrinsic piezoelectric effect

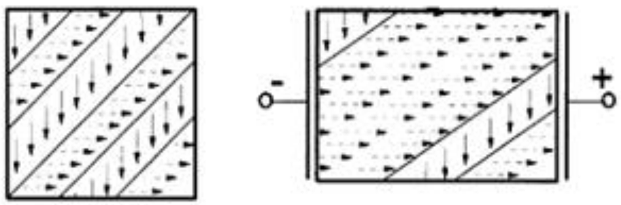
Intrinsic effect – lattice strain

Hoffmann et al. Acta mater. 49 (7), (2001)



The applied electric field causes a shift of the ions in the unit cell, which results in a change of the d-spacing.

Extrinsic effect - texture



The applied electric field leads to domain wall motion which results in an increase or decrease of distinct crystal orientations.

Materials and methods

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- $Ba(Zr_{0.2}Ti_{0.8})O_3$-x$(Ba_{0.7}Ca_{0.3})TiO_3$ (abbreviated as BZT-xBCT),
  x = 0.30 0.32 0.45 0.48 0.52 and 0.60

- Solid state reaction method (M. Acosta):
  Calcination → 2 h at 1300 °C
  Cold isostatic pressing at 300 MPa
  Sintering → 2 h at 1500 ºC

- In-situ electric field investigations: Philips CM20, CM30
- In-situ electric field + cold stage investigations: Tecnai G^2 F20XT

In-situ studies on BZT-xBCT

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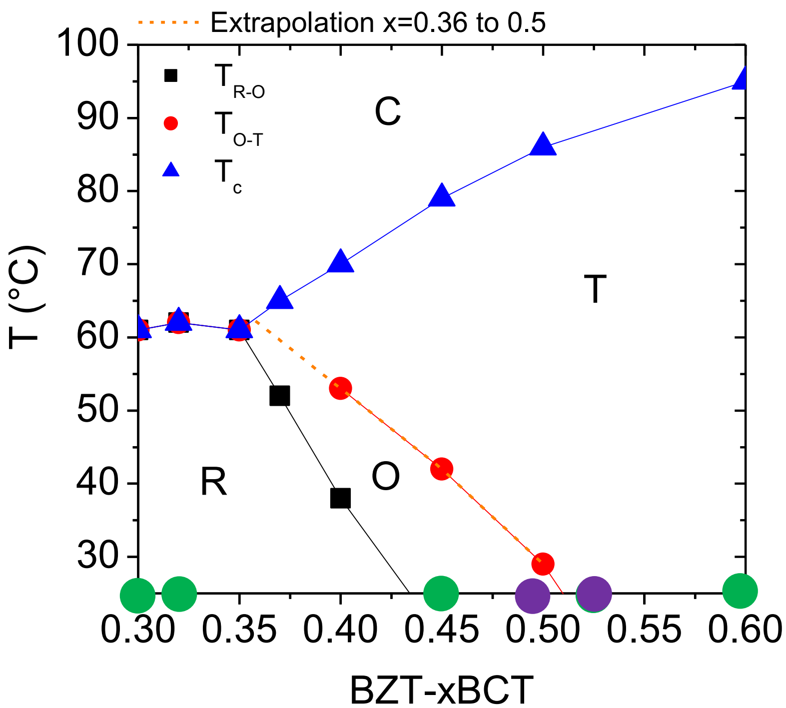
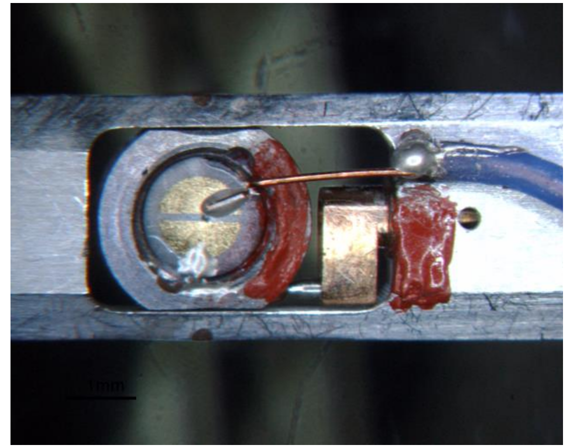
- ● → *In-situ* E-field studies
- ● → *In-situ* E-field + cold-stage studies

x = 0.30, 0.32, 0.45, 0.52, 0.60

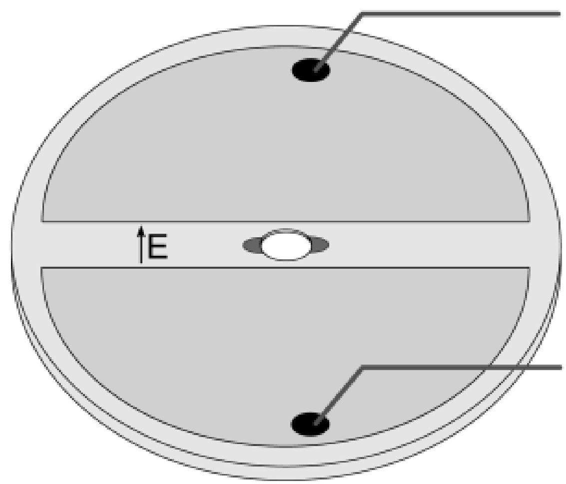
x = 0.48, 0.52





Au-electrodes evaporated on the flat surface

Sample glued into holder with insulating varnish



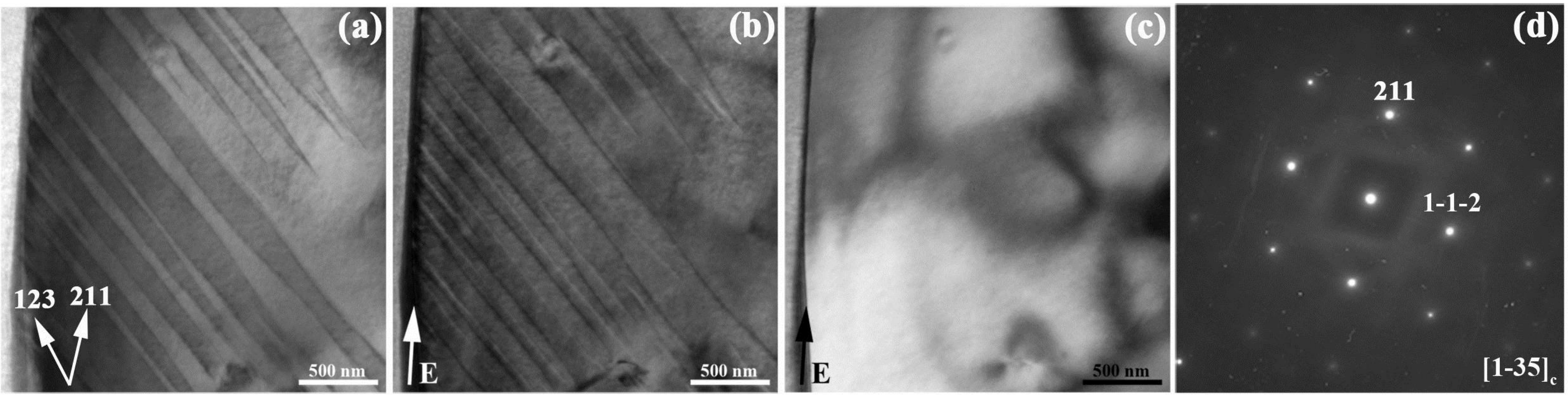
Electrical field perpendicular to electron beam

Distance between electrodes
d = 100µm - 150µm

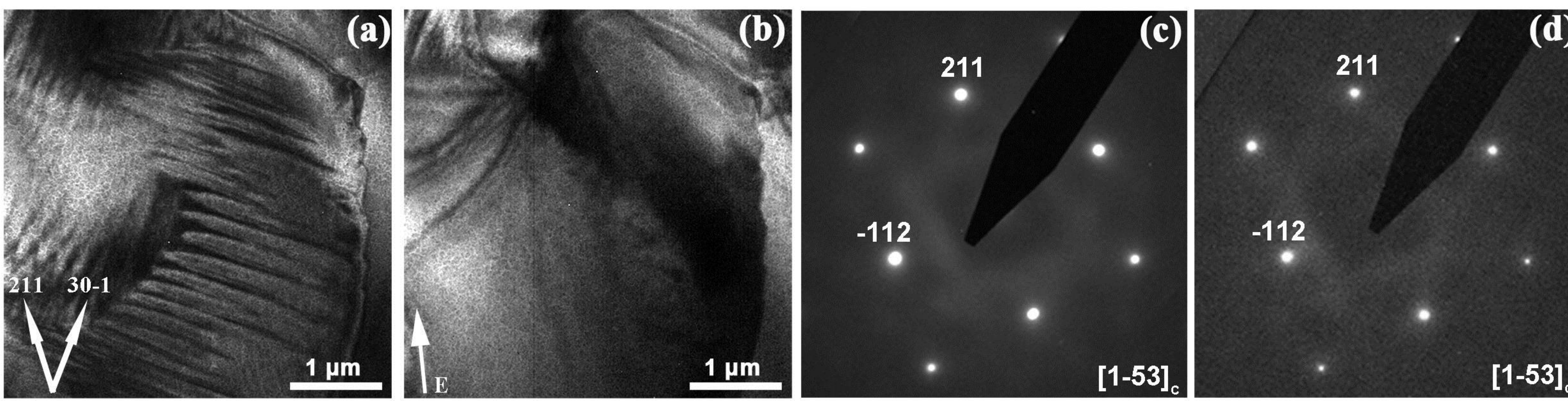
Highest Voltage
U = 500V

In-situ E-field studies BZT-0.30BCT

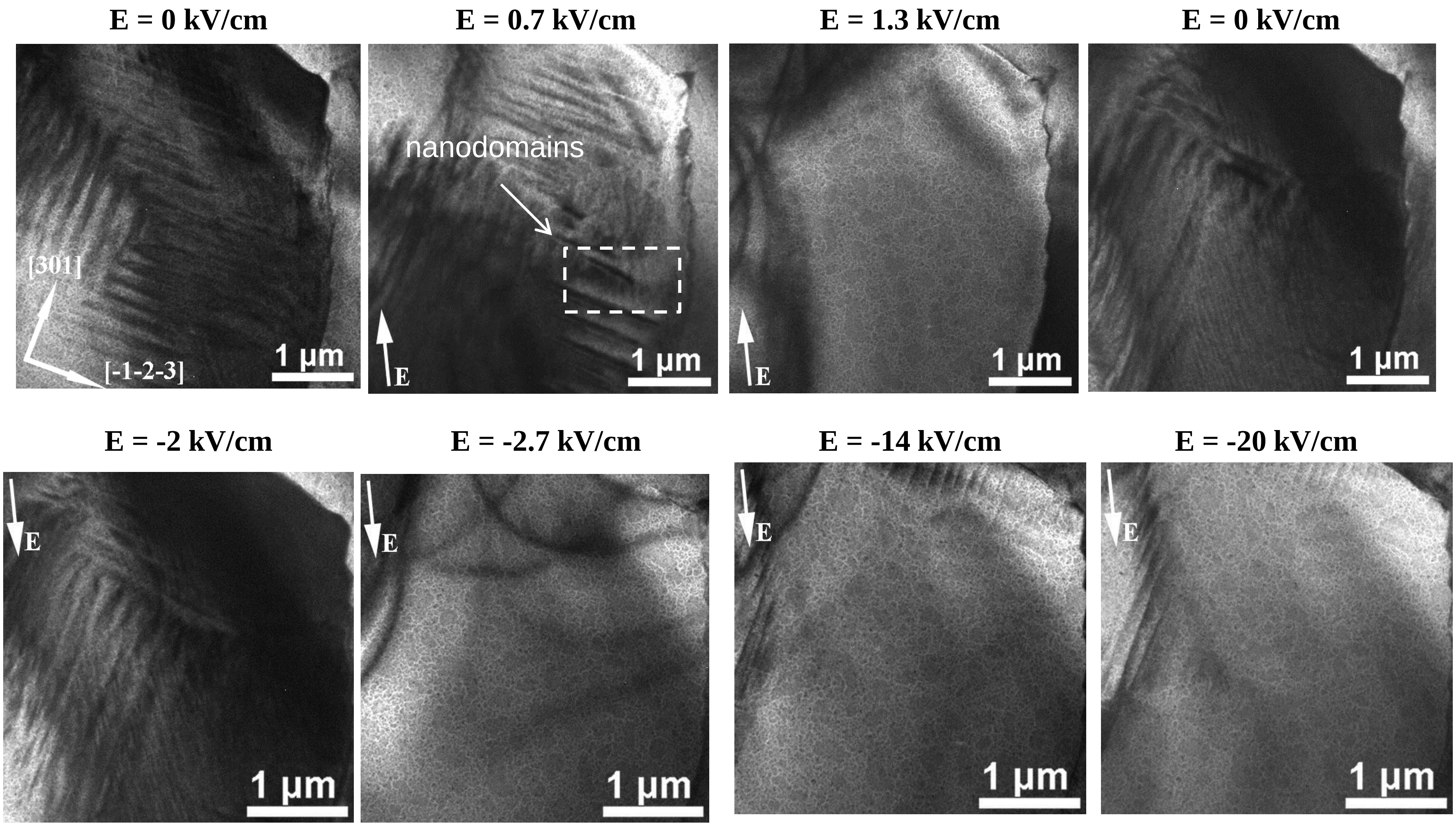
E = 0 kV/cm | E = 3.3 kV/cm | E = 5 kV/cm | E = 0 kV/cm



E = 0 kV/cm | E = 2.7 kV/cm | E = 0 kV/cm | E = 2.7 kV/cm

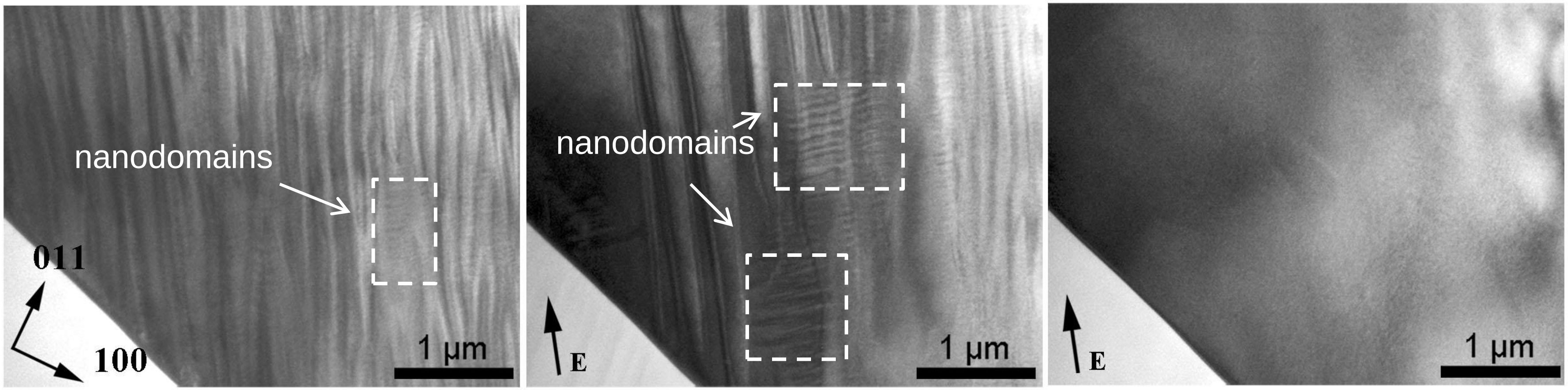


In-situ E-field studies BZT-0.30BCT

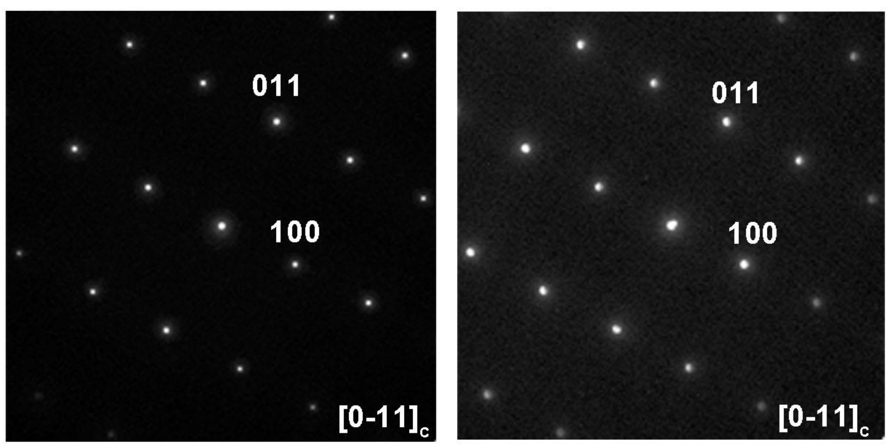


In-situ E-field studies BZT-0.45BCT

E = 0 kV/cm　　E = 2.1 kV/cm　　E = 2.7 kV/cm

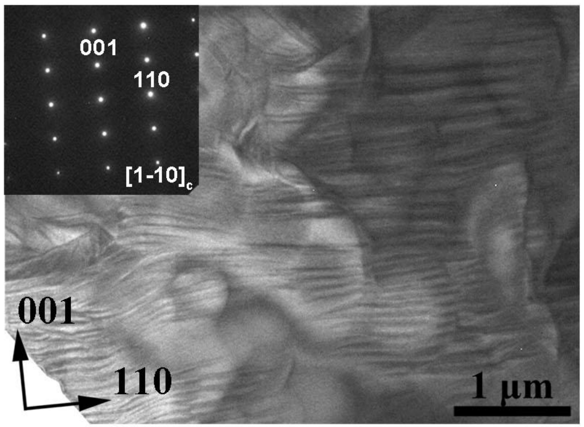


E = 0 kV/cm　　E = 2.7 kV/cm

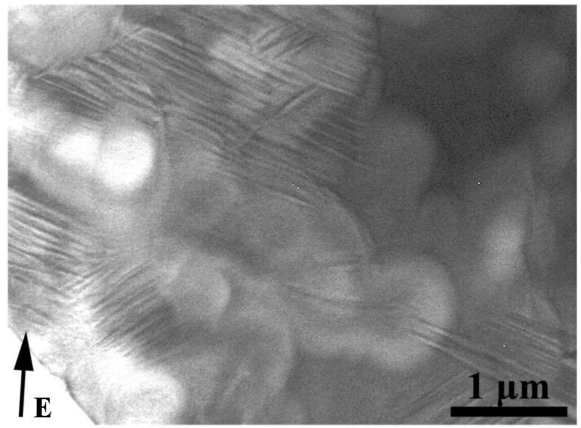


In-situ E-field studies BZT-0.52BCT

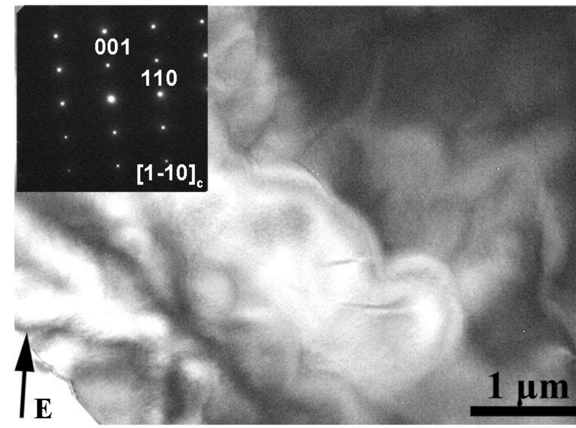
E = 0 kV/cm



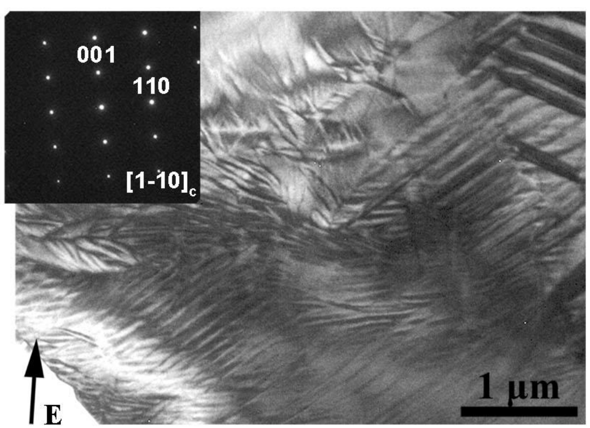
E = 1.25 kV/cm



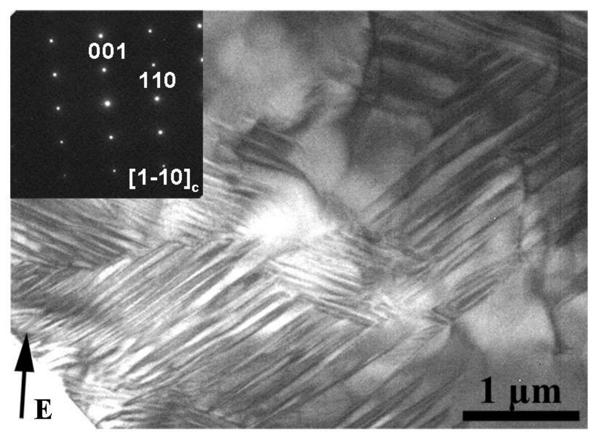
E = 1.7 kV/cm



E = 5.8 kV/cm

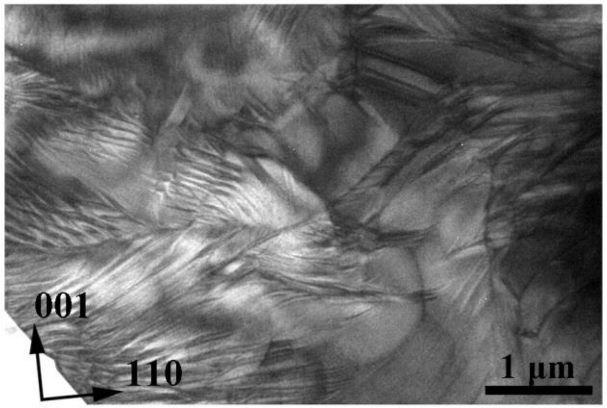


E = 7.5 kV/cm

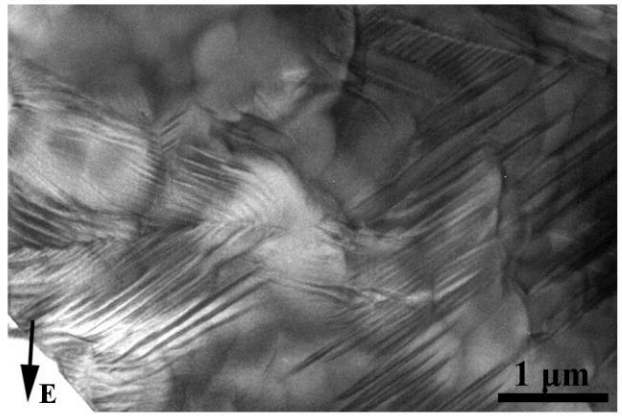


In-situ E-field studies BZT-0.52BCT

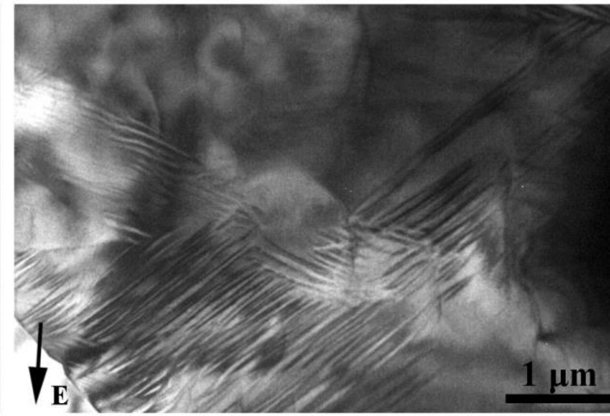
E = 0 kV/cm



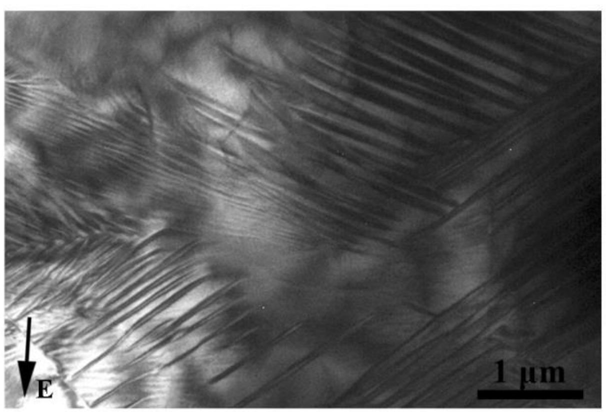
E = -4.6 kV/cm



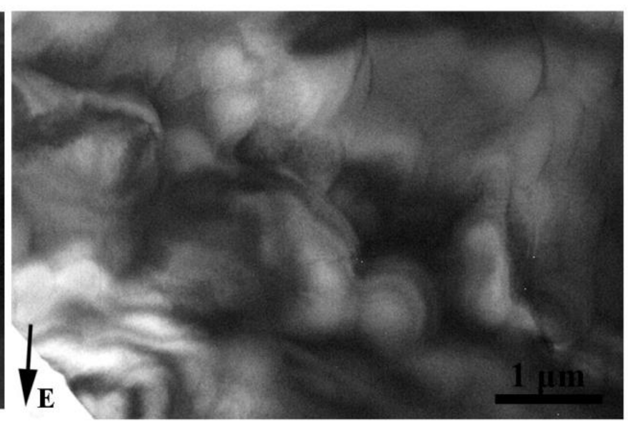
E = -5.4 kV/cm



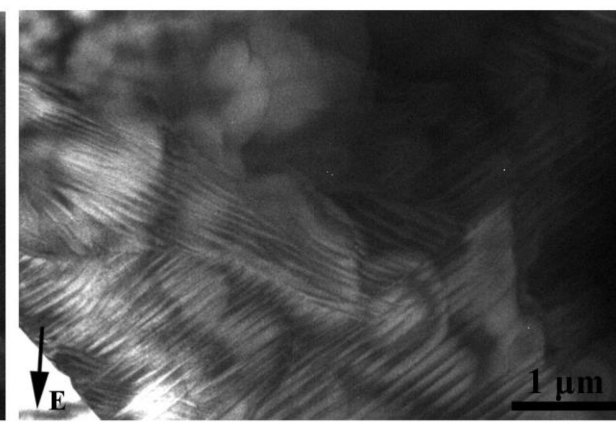
E = -5.8 kV/cm



E = -6.5 kV/cm



E = -6.5 kV/cm after 10 sec



In-situ E-field studies BZT-0.60BCT

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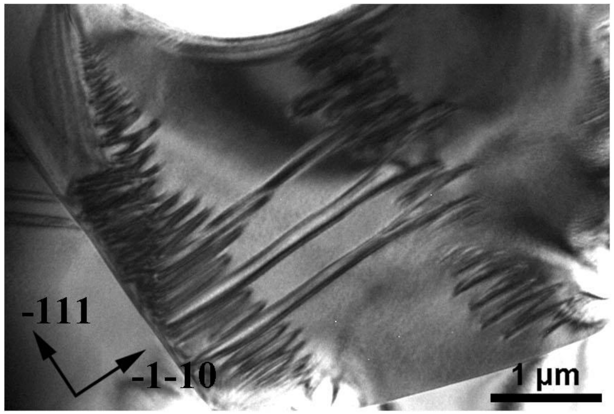
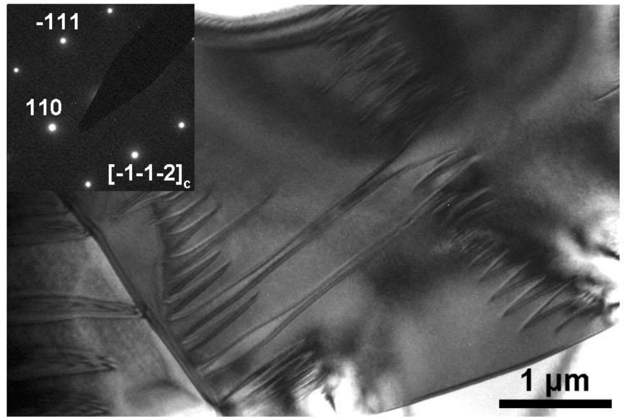
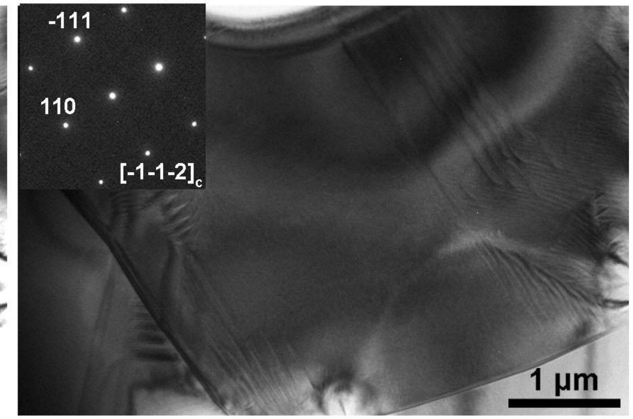
E = 0 kV/cm



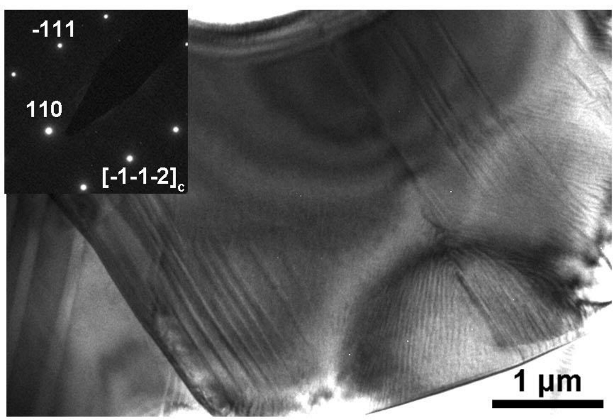
E = 7.66 kV/cm



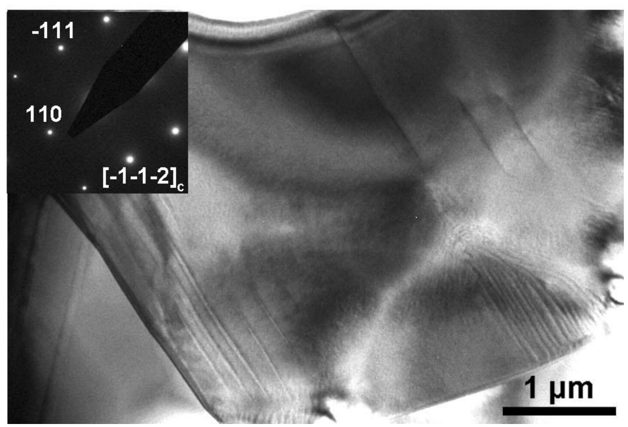
E = 8.66 kV/cm



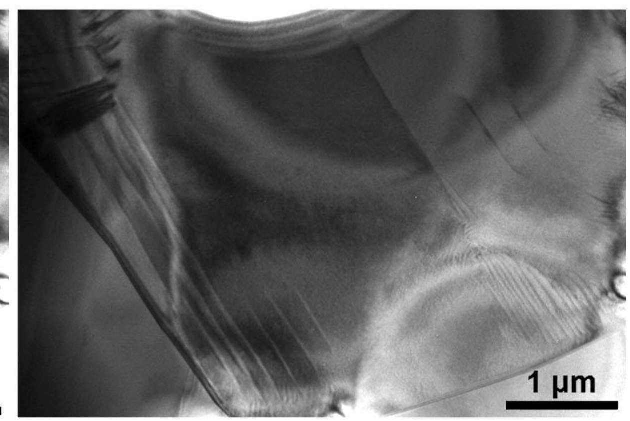
E = 0 kV/cm



E = -7.66 kV/cm



E = -8.66 kV/cm



Scheme of the domain evolution

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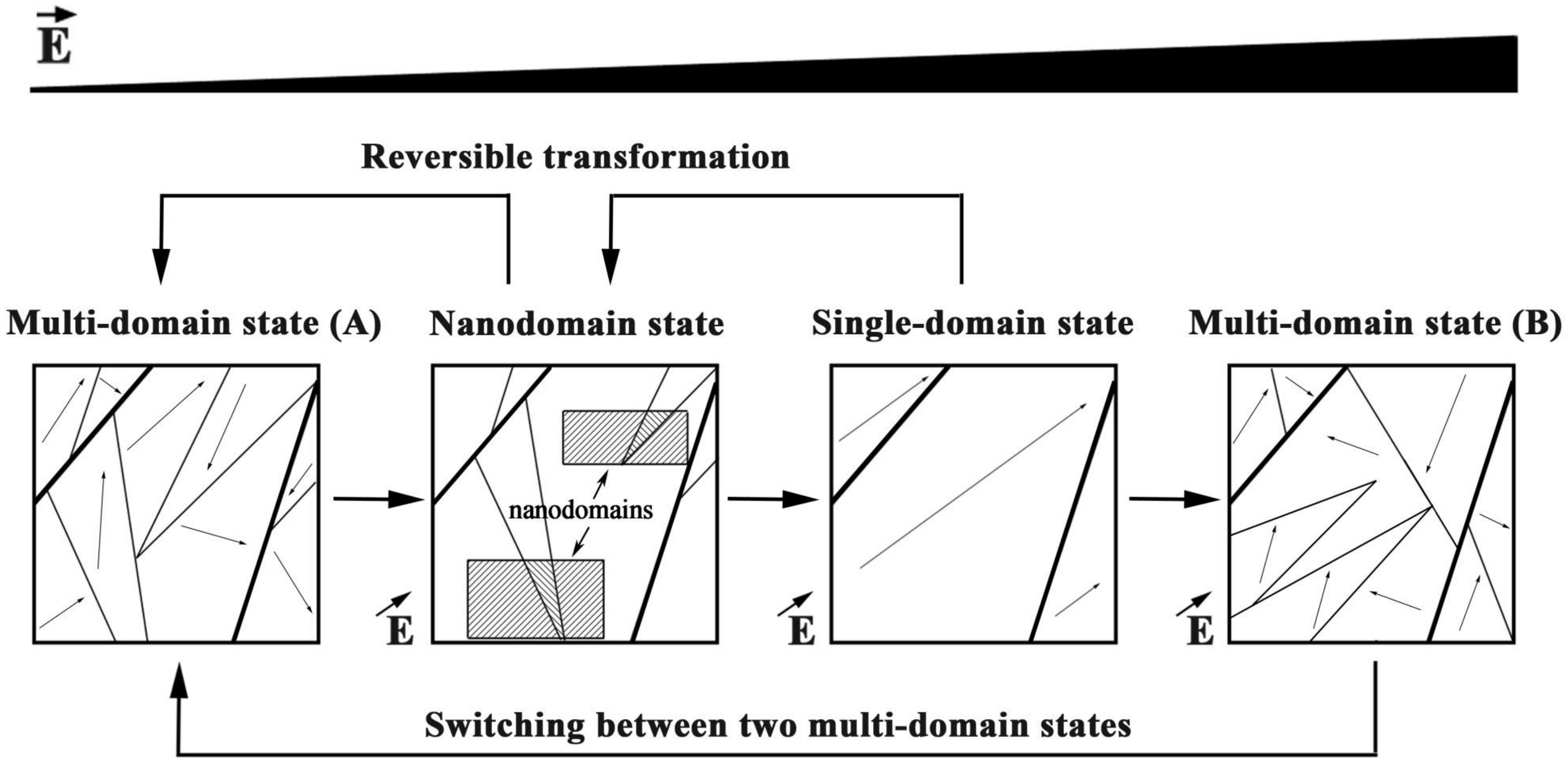

M.Zakhozheva et al. APL, accepted 2014

E-field + cold stage

In situ E-field + cold stage studies BZT-0.52BCT

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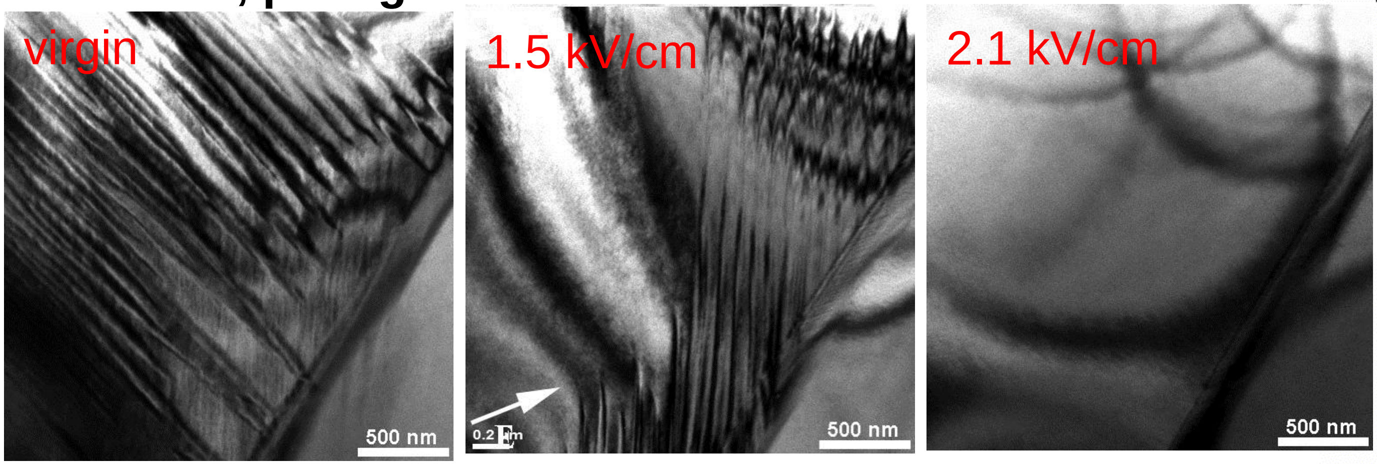
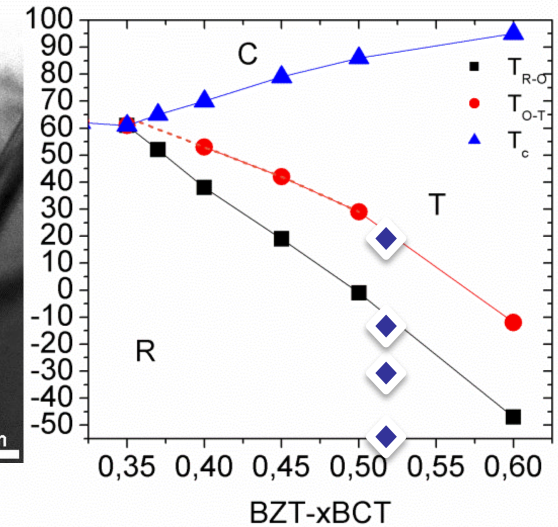
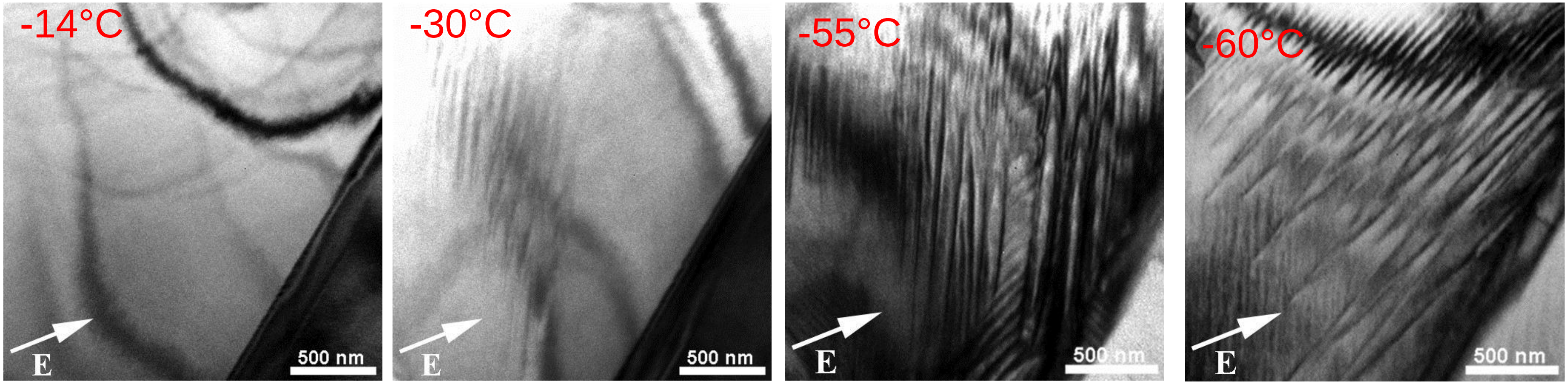
T = 21 °C, poling:





E = 2.1 kV/cm, cool down:



In situ E-field + cold stage studies BZT-0.48BCT

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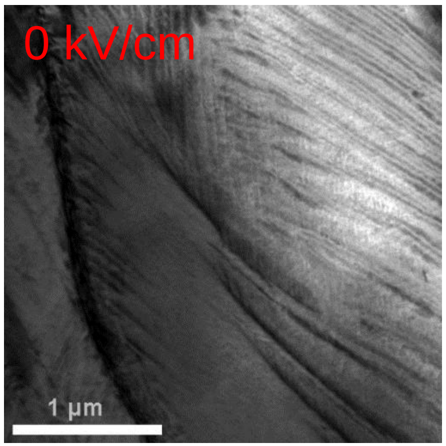
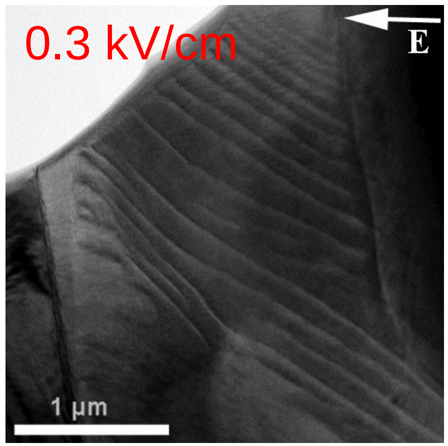
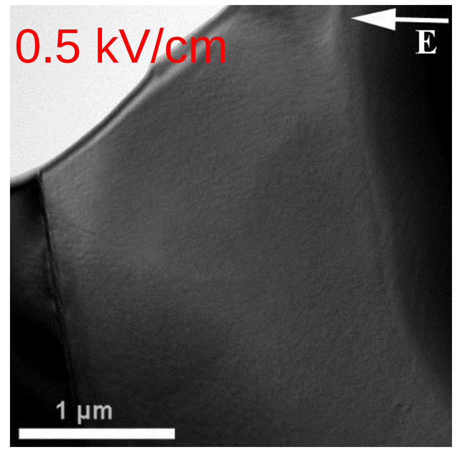
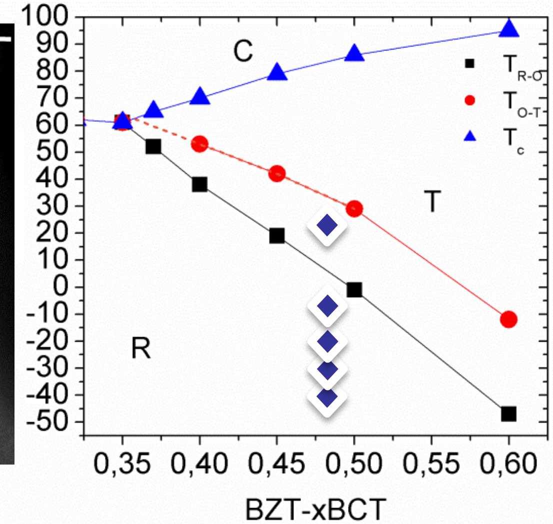
T = 21 °C, poling:

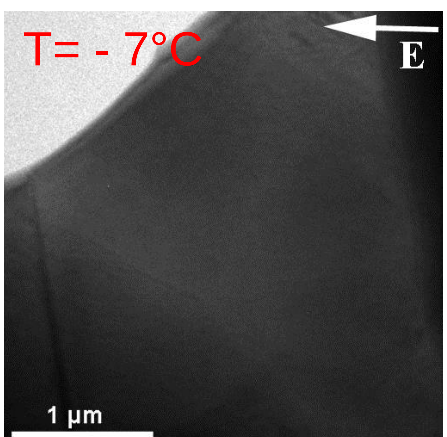


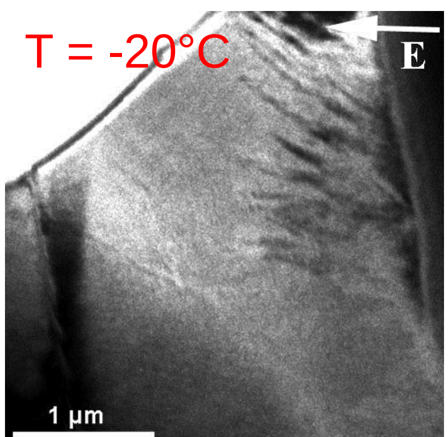


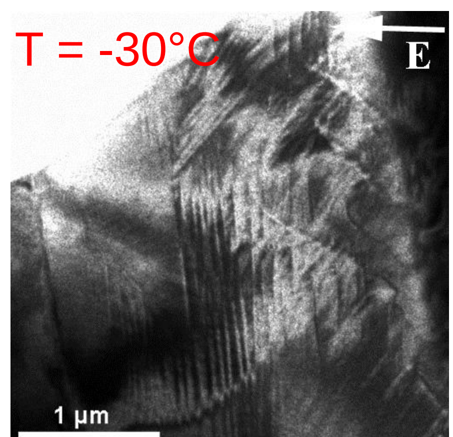


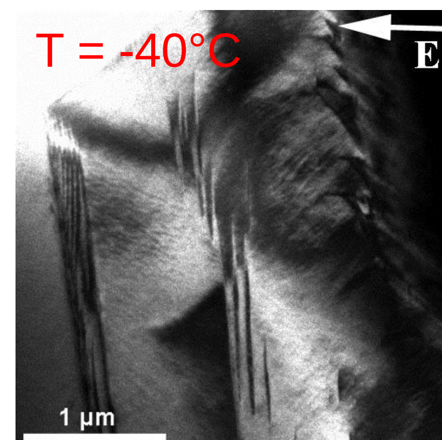


E = 0.5 kV/cm, cool down:









In situ E-field + cold stage studies BZT-0.48BCT

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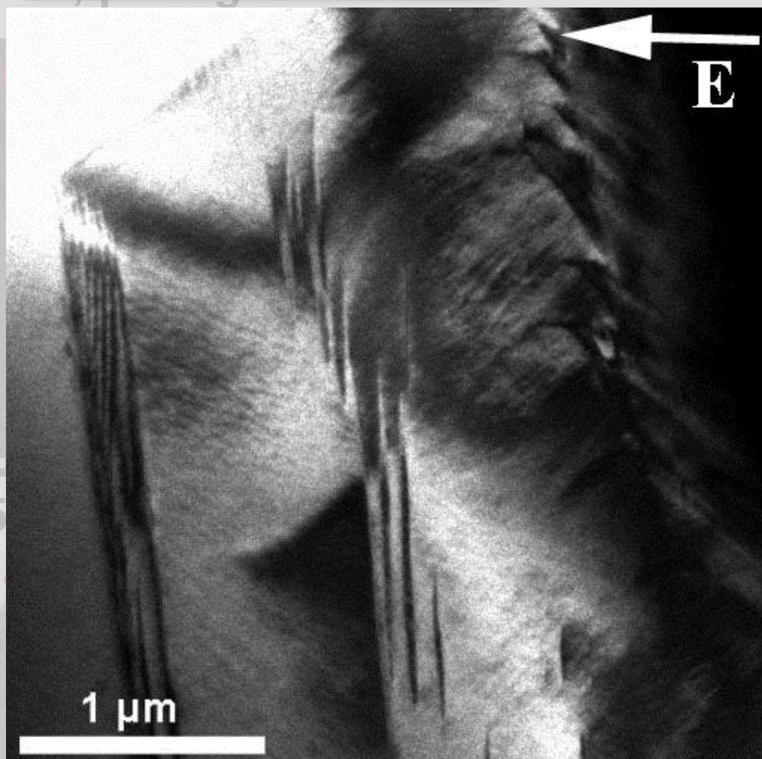
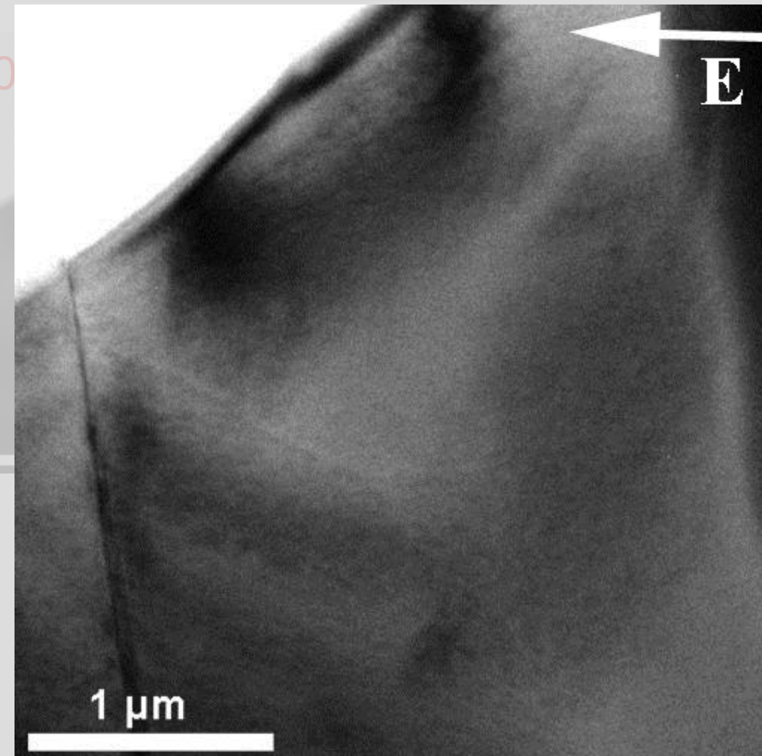
T = -40°C
E = 0.5 kV/cm



T = -40°C
E = 0.6 kV/cm



Summary

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In the present study, a field-induced transformation from a multi- to a single-domain state was monitored in $Ba(Zr_{0.2}Ti_{0.8})O_3$-x$(Ba_{0.7}Ca_{0.3})TiO_3$ piezoceramics.

- Transformation from the multi- to the single-domain state with increasing the poling field.
- Appearance of the nanodomain state at the moderate poling fields.
- Single-domain state is not stable against higher fields.
- Transformation from the single-domain state to the multi-domain state with decreasing the temperature at the constant poling field
- SAED patterns do not show any reflection splitting or any detectable changes during the poling process.

Conclusion

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The displacement of the domain walls and changes in the domain configuration during poling indicated a high extrinsic contribution to the piezoelectric properties in $Ba(Zr_{0.2}Ti_{0.8})O_3$-x$(Ba_{0.7}Ca_{0.3})TiO_3$ piezoceramics.

Thank you for your attention