Challenges and New Trends for Piezoelectric Actuators

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Polycrystalline lead zirconate titanate (PZT) has been the material of choice for piezoelectric applications such as medical ultrasound, sonar, fuel modulation, and active vibration damping; however, there have been no significant improvements in properties in recent years in this now mature material system. The main focus in the piezoelectric material development beyond the capabilities of PZT has been two-fold: (i) increasing the electromechanical activity, and (ii) increasing the operating temperature.

(i) Remarkable properties have been reported in recent years for an emerging class of piezoelectric crystals based on lead magnesium niobate (PMN) substituted with lead titanate (PT). These new material systems can have exceptionally large piezoelectric coefficients and electromechanical coupling factors (e.g., k_{33}=0.94). The latter implies over 90 % electro-mechanical energy conversion (k^2), and the former are approximately an order-of magnitude greater than commercially available piezoelectrics based on polycrystalline PZT. Recently, developments in the growth of PMN-PT single crystals and discovery of a unique direction among the crystallographically equivalent axes have made it feasible to use single crystal elements in high-end piezoelectric applications.

(ii) Development of high temperature piezoceramics has been a tougher challenge to meet. Most high temperature piezoelectric material systems suffer from low piezoelectric coefficients and are only suitable for sensor applications. Two main challenges in producing high temperature piezoelectrics are (a) to increase Curie temperature (T_C) without an increase in loss tangent, and (b) to demonstrate high piezoelectric activity. $BiScO_3$-$PbTiO_3$ (BS-PT) based systems exhibited high T_C and large piezoelectric coefficients near the morphotropic phase boundary (MPB). The effects of excess PbO and Bi_2O_3 and their partitioning in grain boundaries were studied using impedance spectroscopy, ferroelectric, and piezoelectric measurement techniques. Excess Bi_2O_3 proved to be a successful liquid phase forming additive to improve the BS-PT piezoceramics for high temperature applications, as a result of increased resistivity and enhanced piezoelectric activity through microstructure engineering.

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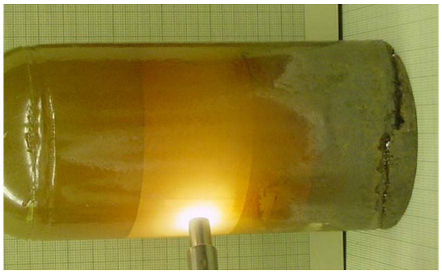

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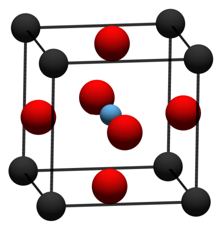
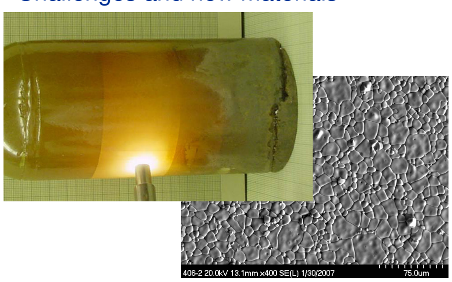


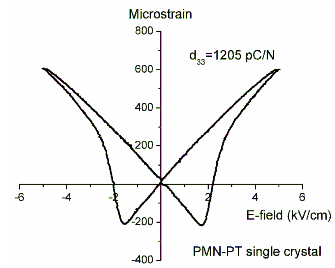
Case Western Reserve University est. 1826

Outline

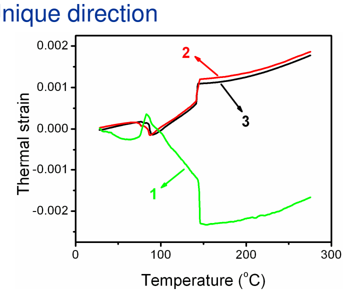
Challenges and new materials



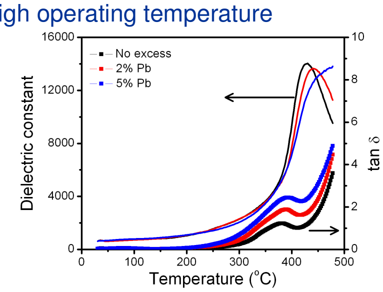
High piezoelectric coefficient



Unique direction



High operating temperature



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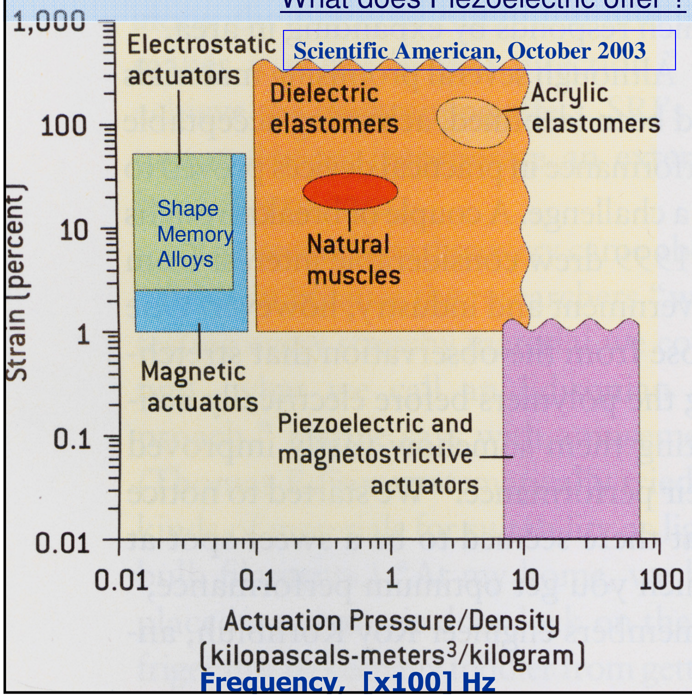
What does Piezoelectric offer ?



COMPARISON:

- **Same muscle tissue powers both insects and elephants. But, muscle but does not work at high temperatures and frequencies.**
- **Gas turbine applications require high frequency and displacement at elevated temperatures.**

Poling

All materials (32)

⇓

Piezoelectrics (20)
Non-centrosymmetric

⇓

Pyroelectrics (10)
Polar

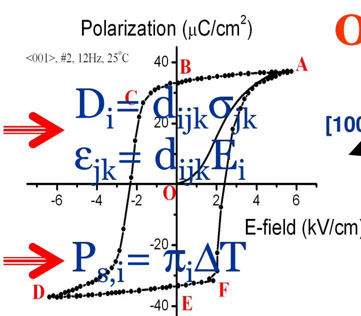
⇓

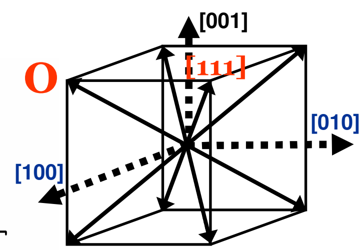
Ferroelectrics
Reorientable domains

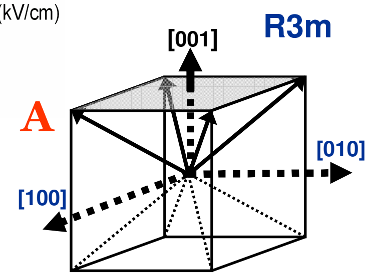
$D_i = d_{ijk}\sigma_{jk}$

$\varepsilon_{jk} = d_{ijk}E_i$

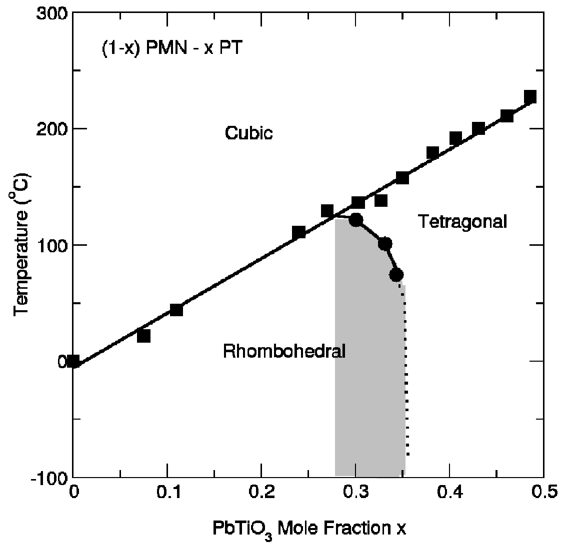
$P_{s,i} = \pi_i \Delta T$







PMN-PT



Shrout T., Zung P. C., Namchul K., Markgraf S. Ferroelectrics Letters **12**: 63-69, 1990.

- Remarkable properties in the MPB region.

Crystal numbers

1 ⟶ 6

Critical point (CP) — MPB

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Dielectric properties of <001> poled crystals

Specimen	Dielectric properties at 25 °C				Transition temperature (°C)	
Specimen	K'_{33}	tan δ	d_{33} (pC/N)	k_{33}	R→T	T→C
1	5000	0.002	1200	0.914	104	112
2	5625	0.002	1475	0.920	105	113
3	6025	0.003	1625	0.927	105	120
4	8465	0.002	3470	0.934	97	129
5	9195	0.004	3620	0.939	97	132
6	14260	0.005	4080	0.951	87	137
PZT 5H	3400	0.025	590	0.75	195	

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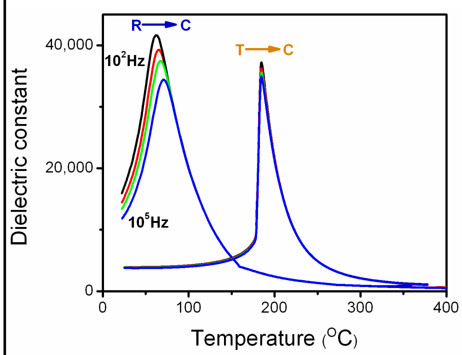
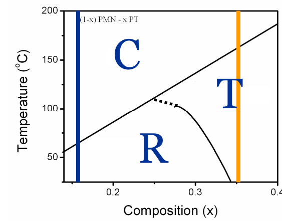
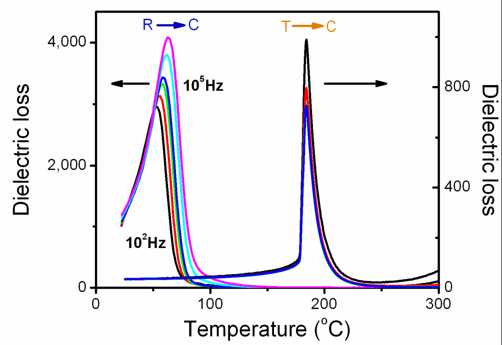
A. Sehirlioglu et al., J. Appl. Phys. **99**, 064101 (2006).

Outside the MPB region, <001> direction

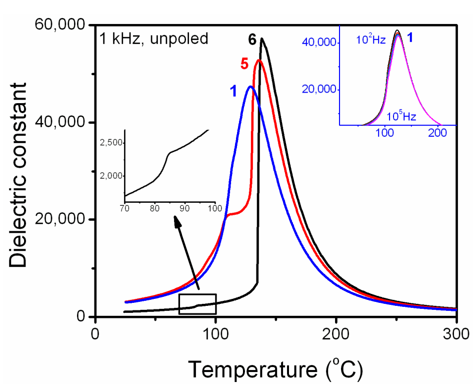


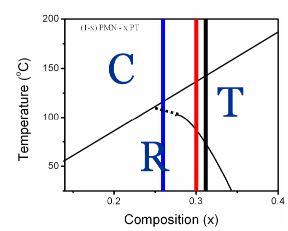


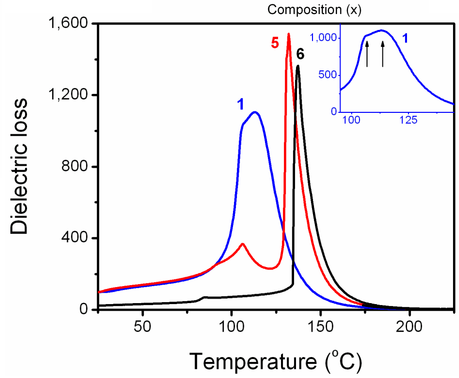


Unpoled

In the MPB region, <001> direction

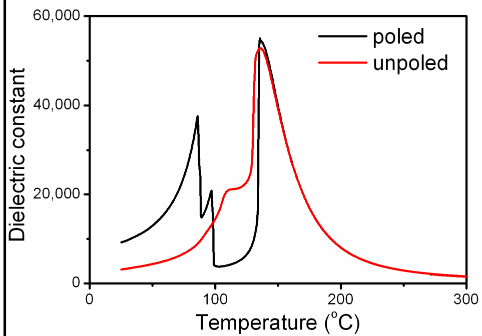




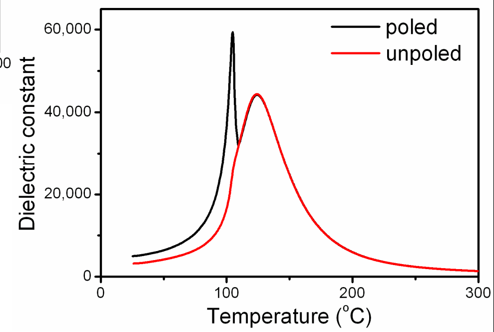


Diffuse transformation is not due to relaxor behavior

Effect of poling, <001> direction



Close to the CP



Close to the MPB

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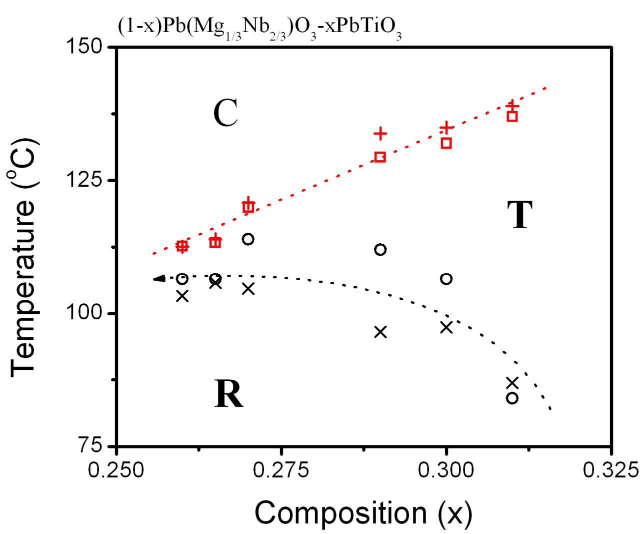
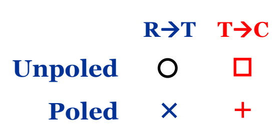
Effect of poling on phase transformation temperatures



Poling along <001>



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Summary (Dielectric behavior)

- Diffuse K_{33} vs. T transformation without frequency dispersion
  (convergence of R→T→C phases)
- Effects of poling along <001>
  - increases K_{33} at room temperature
  - decreases the R→T transformation temperature
  - increases K_{33} at R→T
- Poling does not have any effect on the dielectric behavior of the T→C phase transformation.

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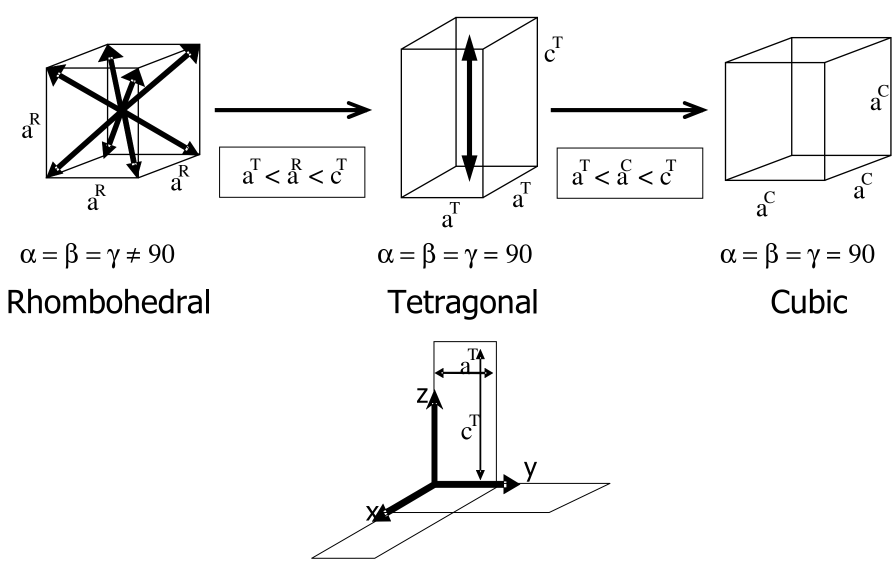
Domain orientations, unpoled



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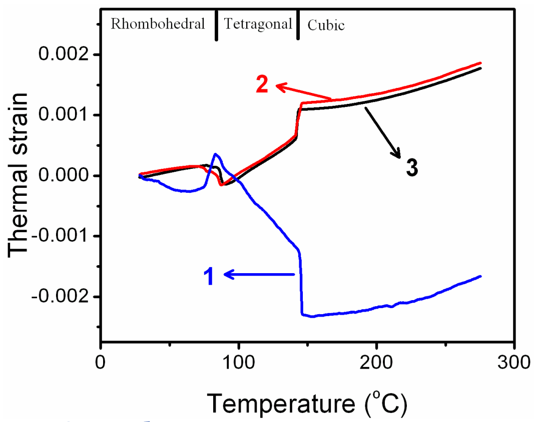
Effect of orientation, <001>, unpoled

All along <001>



2°C/min, heating

	Along 1 (ppm/K)	Along 2 and 3 (ppm/K)
α^R	-8.6	4.5
α^T	-25	14
α^C	17	17
K_{33}	5424	1765 1956

Unpoled

A. Sehirlioglu et al., Phys. Rev. B **72**, 214110 (2005)

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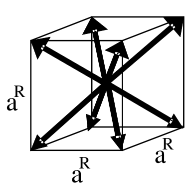
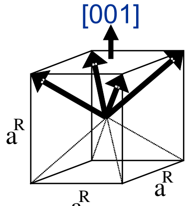
Effect of poling – measured parallel to poling

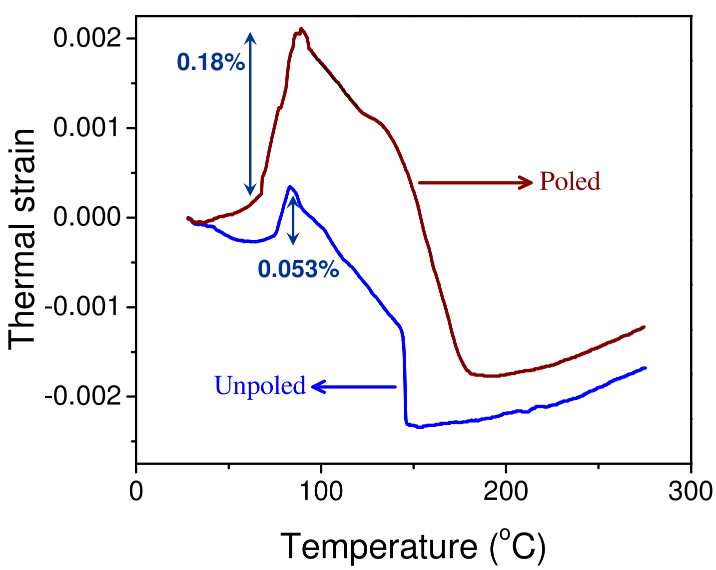
Unpoled



Poled

[001]





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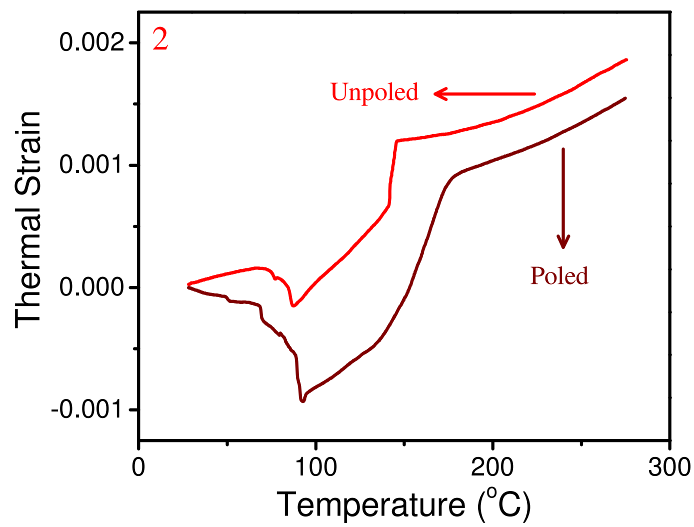
Effect of poling - measured perpendicular to poling direction



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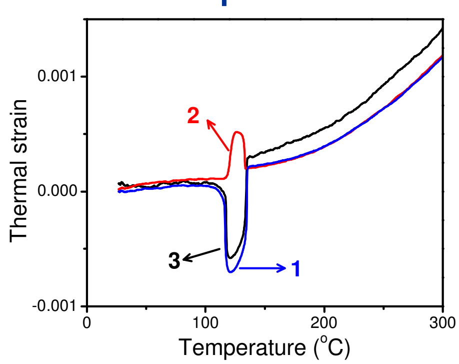
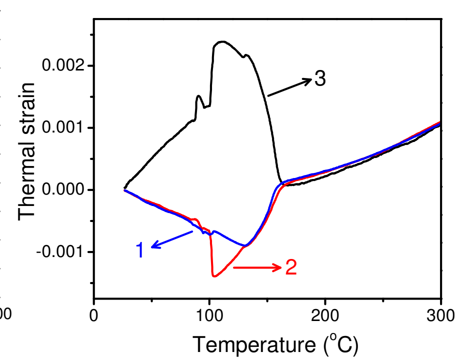
Effect of poling (near critical point)

Unpoled



Poled along 3



- Poling reorients the unique direction
- α^R shows uniaxial macroscopic symmetry
- Depoled is the same as unpoled

K_{33} =8430 (poled parallel)

K_{33} =4573 (poled normal)

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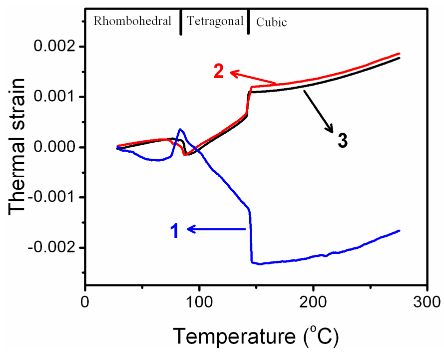
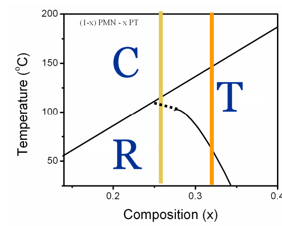
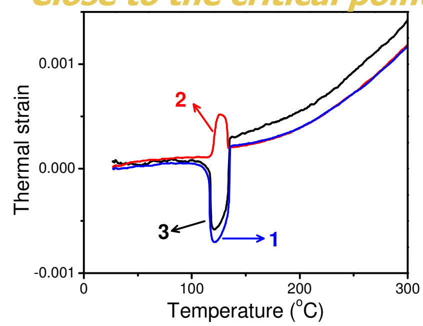
Effect of composition (unpoled crystal)

Close to the MPB





Close to the critical point



K_{33}	Unique	Normal 1	Normal 2
MPB	5425	1955	1765
Critical point	4870	4710	4685

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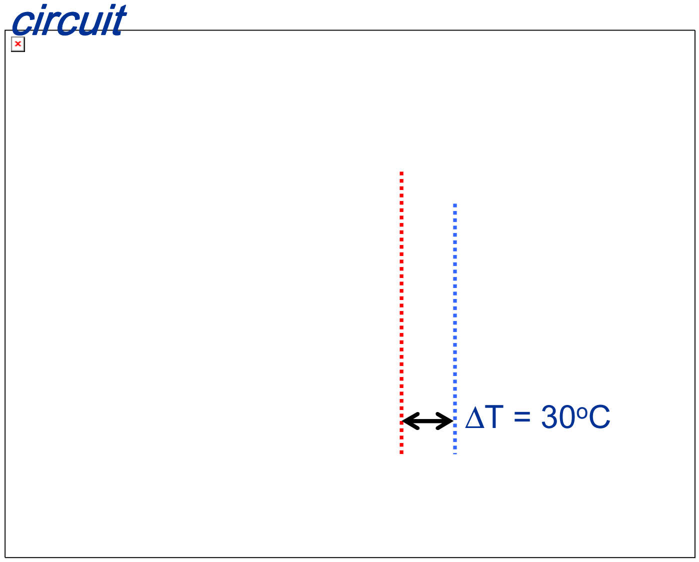
Effect of poling on T→C temperature

Open circuit



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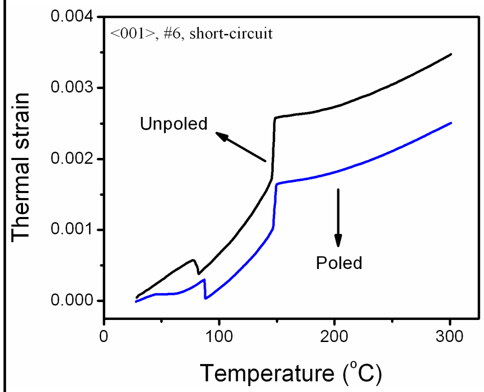
Boundary conditions



Short circuit
(ΔE=0)

Open circuit
(ΔP=0)

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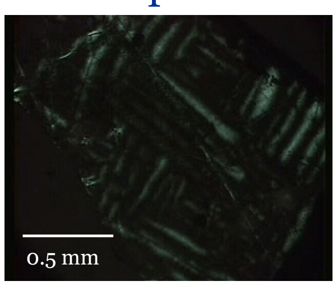
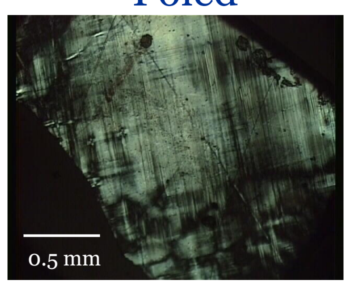
T→C phase transformation

Unpoled	Poled

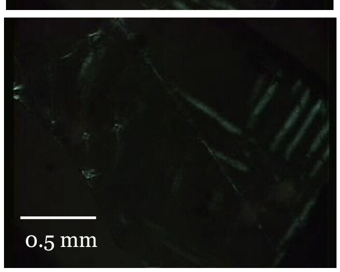
141°C

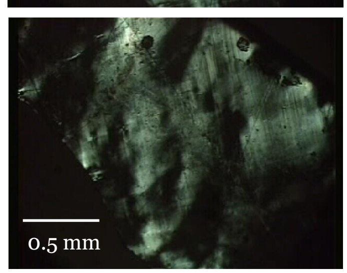




142°C

142°C





150°C

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Summary

- not all of the <001> directions were macroscopically equivalent even though they were *indistinguishable* by XRD.
- the anomaly may be attributable to the method of seeded crystal growth.
- The unique direction was related to the domain structure of the crystal and could be reoriented by room temperature poling in a direction normal to the unique direction.
- Highest K is obtained when poled along the unique direction
- Poling promoted the formation of *c*-oriented tetragonal domains along the poling direction.
- the phase transformations were sharper under short-circuit boundary conditions.

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Objective

Development of high-temperature piezoelectric actuators for aeronautics and aerospace applications.

Applications

- Actuators for Aerospace and Aeronautics
  - Fuel modulation, valves, micro-positioning devices, MEMS, active damping and energy harvesting.
- Sensors
  - Pressure sensors, passive damping

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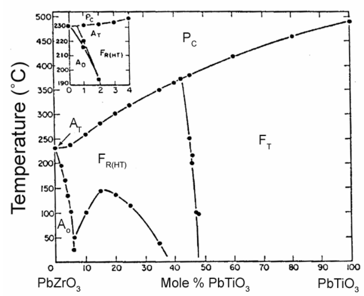
Challenges for High Temperature

- Trade off between T_C and d_{33}
- Conductivity at elevated temperatures

	T_C (°C)/(°F)	d_{33} (pC/N)
PZT Type II (PZT 5A)	350 / 662	374
PMN-PT single crystals	90 / 194	>2000
$BiScO_3$-$PbTiO_3$	450 / 842	401
$La_3Ga_{5.5}Ta_{0.5}O_{14}$ single crystal	N/A	7
$Na_{0.5}Bi_{4.5}Ti_4O_5$	650 / 1202	19
$La_2Ti_2O_7$	1482 / 2700	16



B. Jaffe, W. R. Cook and H. Jaffe, Piezoelectric Ceramics, Academic Press, New York, 1971.

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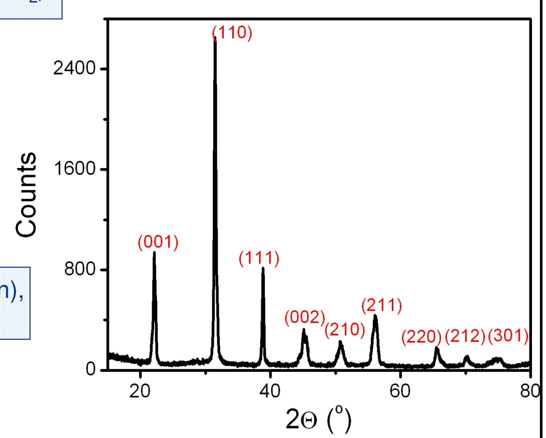
Processing of BS-PT

Raw materials (Bi_2O_3, PbO, Sc_2O_3, TiO_2)
→ Ball milling (15hrs)
→ Drying (stirred)
→ Calcination (750°C, 3hrs, 5°C/min), in air
→ Ball milling (6hrs) ← Excess addition
→ Pressing
→ Sintering (1100°C, 1hr, 5°C/min), in air



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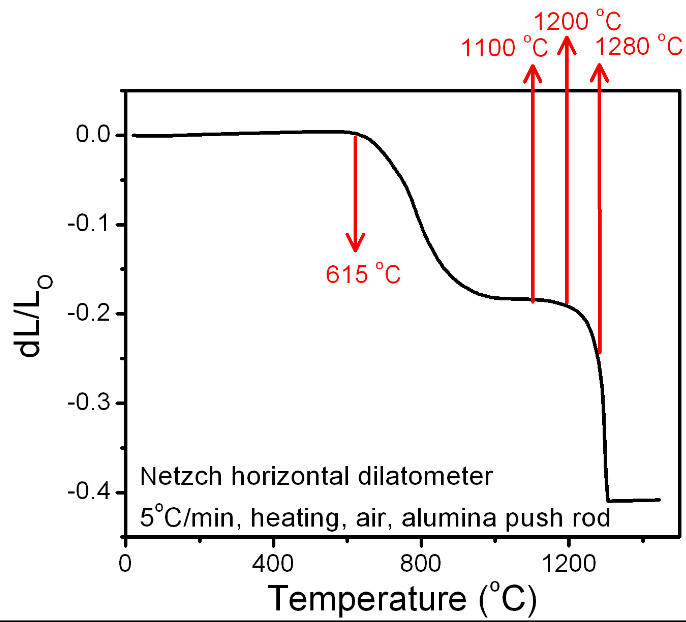
Sintering Conditions for BS-PT



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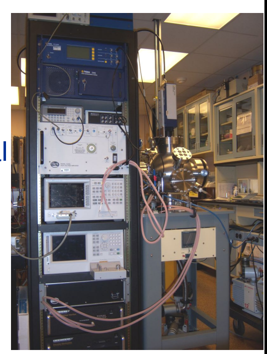
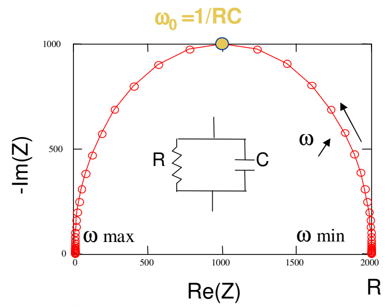
Electrical characterization

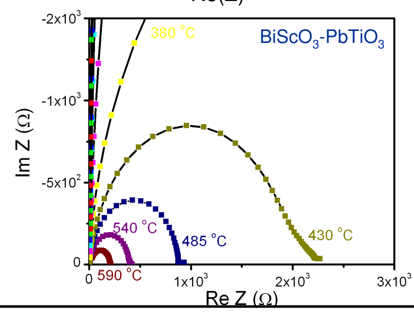


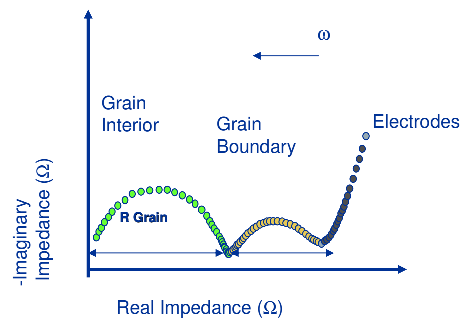
- Impedance measurements *(Solartron and HP Agilent)*
  - 1Hz-1MHz, Room temperature to 1000 °C.
  - 40Hz-110MHz, Room temperature to 600 °C.
  - 1Mhz-3Ghz, Microwave range
    (Determination of electrical, dielectric and electromechanical properties)
- Ferroelectric measurements *(Radiant Technologies)*
  - Bipolar, unipolar loops, leakage (up to 10,000V)
- Piezoelectric measurements
  - Laser dopplermeter *(Polytech)* coupled with a signal generator and a high power amplifier (up to 10,000V)
  - PhotonicTM sensor *(MTI technologies)* coupled with Radiant
  - Berlincourt d_{33} -meter

Impedance Spectroscopy

Conductivity - Grain/Grain Boundary







R // C : Resonance frequency ω_0 =1/RC

conduction: grain ≠ grain boundary

ω_0 grain boundary << ω_0 grain-Low T

ω_0 grain boundary ~ ω_0 grain-High T

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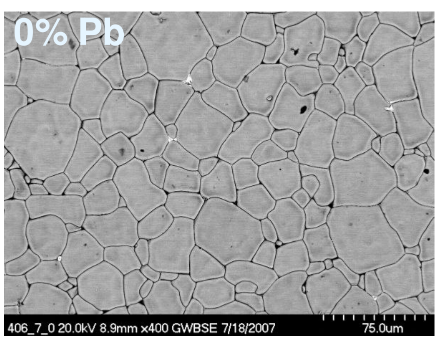
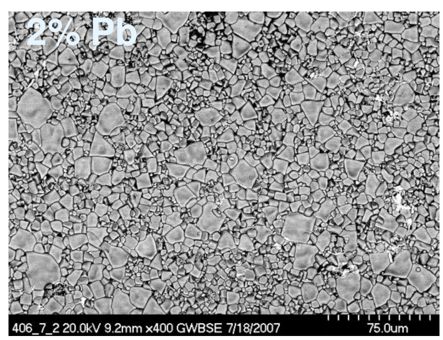
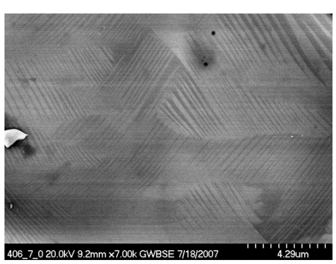
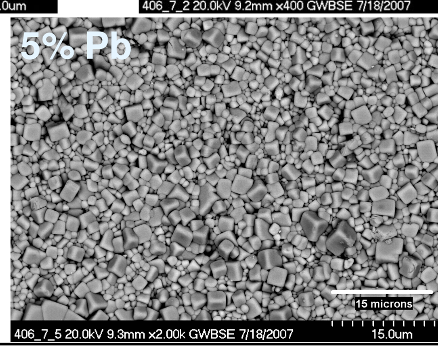
Effect of Pb on microstructure







0% Pb



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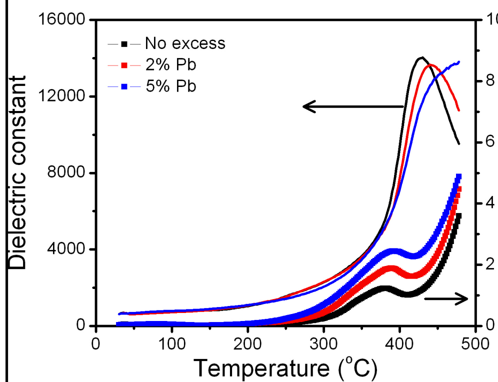
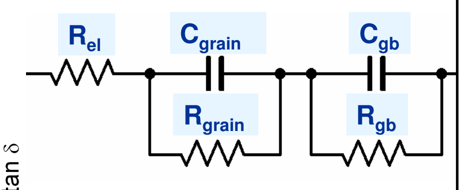
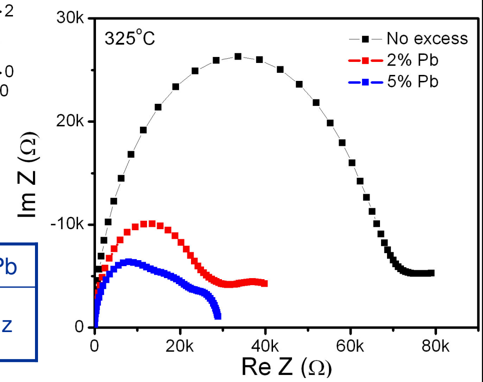
Effect of Pb in BS-PT





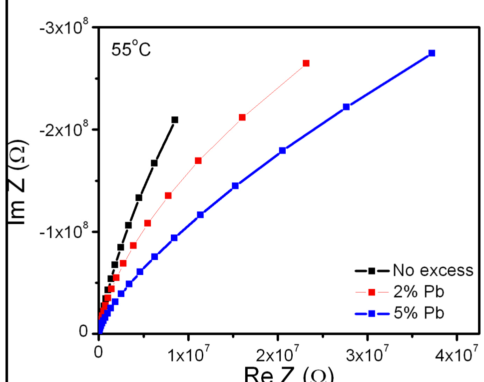
10 kHz, 0.5 V/mm ac, in air

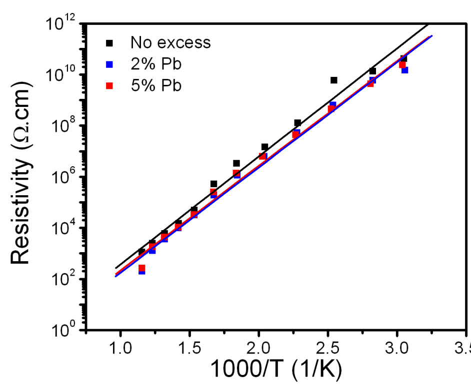
Grain boundary contribution



	0% Pb	2% Pb	5% Pb
Relaxation frequency	800 Hz	3 kHz	5 kHz

Effect of Pb in BS-PT





	ρ_{grain} (Ω.cm)	Dielectric constant, K_{grain}	E_{ac} (ev)
No excess	4.1×10^{10}	1085	0.36
2%Pb	2.4×10^{10}	805	0.35
5%Pb	1.4×10^{10}	727	0.35

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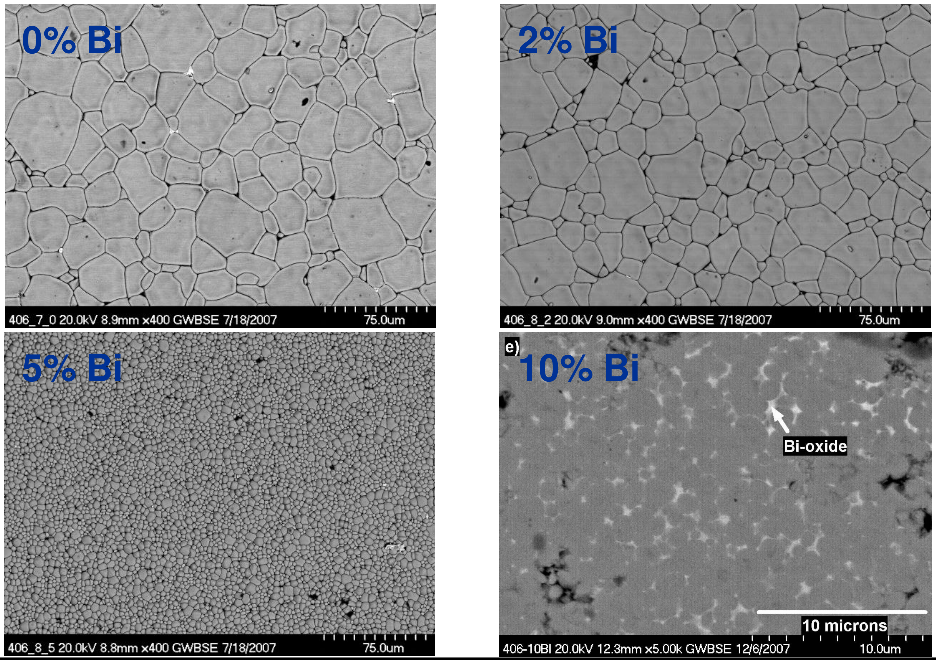
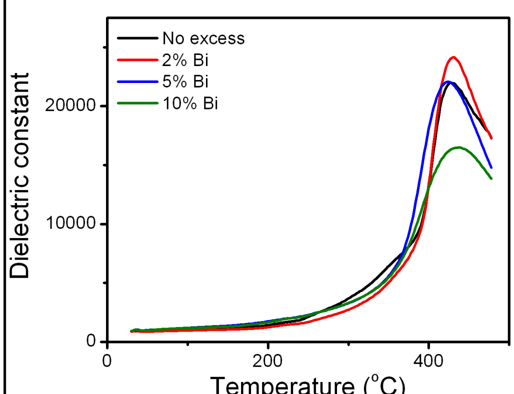
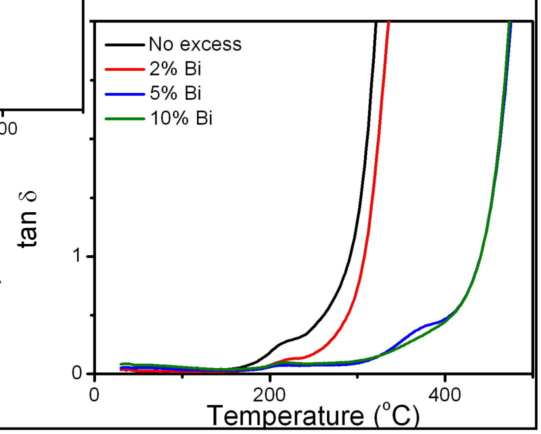
Effect of Bi on microstructure



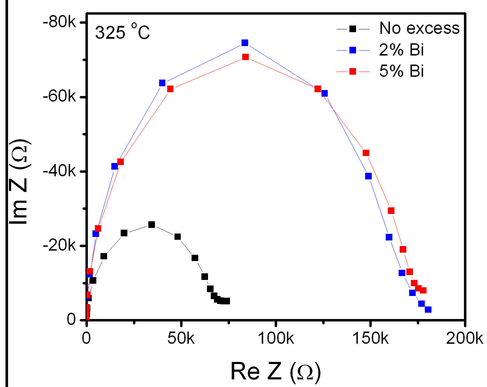
Effects of Bi in BS-PT

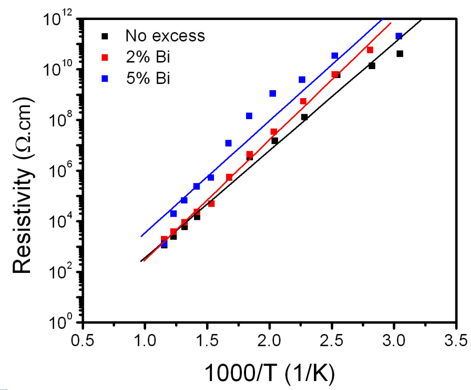


1 kHz, 0.5 V/mm ac, in air



Effect of Bi in BS-PT





	ρ_{grain} (Ω.cm)	Dielectric constant, K_{grain}	Relaxation freq.	E_{ac} (ev)
No excess	4.1×10^{10}	1085	800 Hz	0.36
2% Bi	4.9×10^{10}	1104	400 Hz	0.40
5% Bi	19.6×10^{10}	1402	200 Hz	0.38

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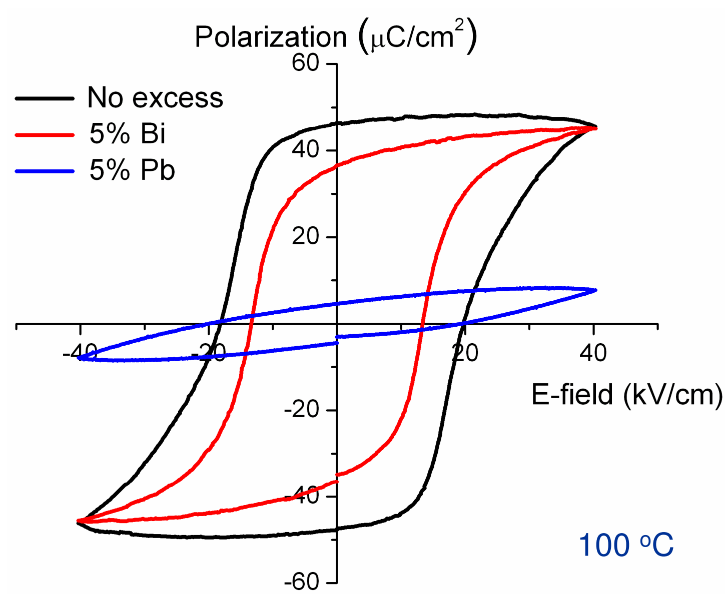
Ferroelectric and piezoelectric properties



100 °C

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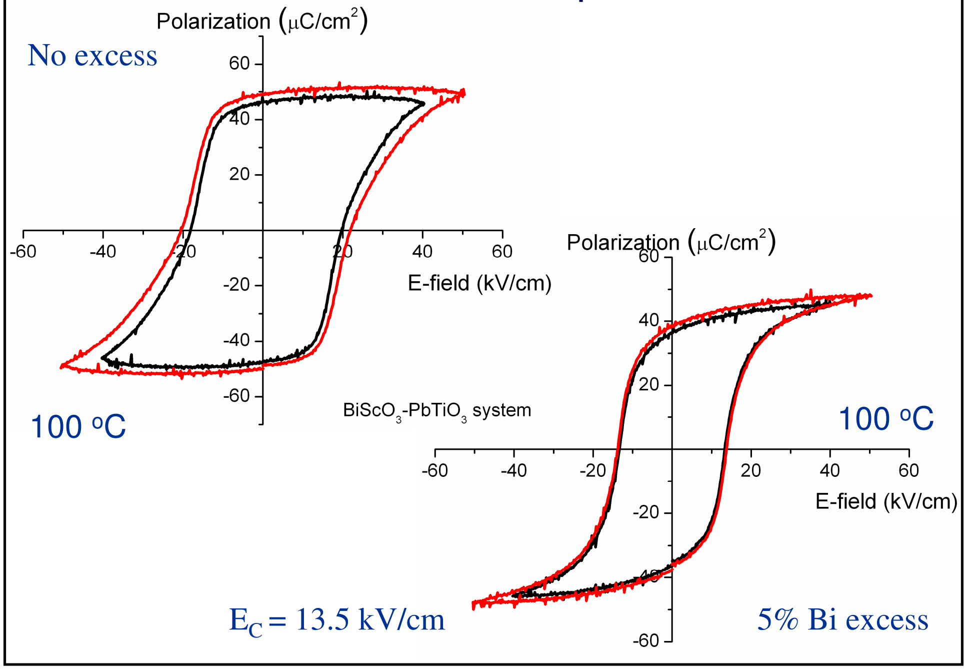
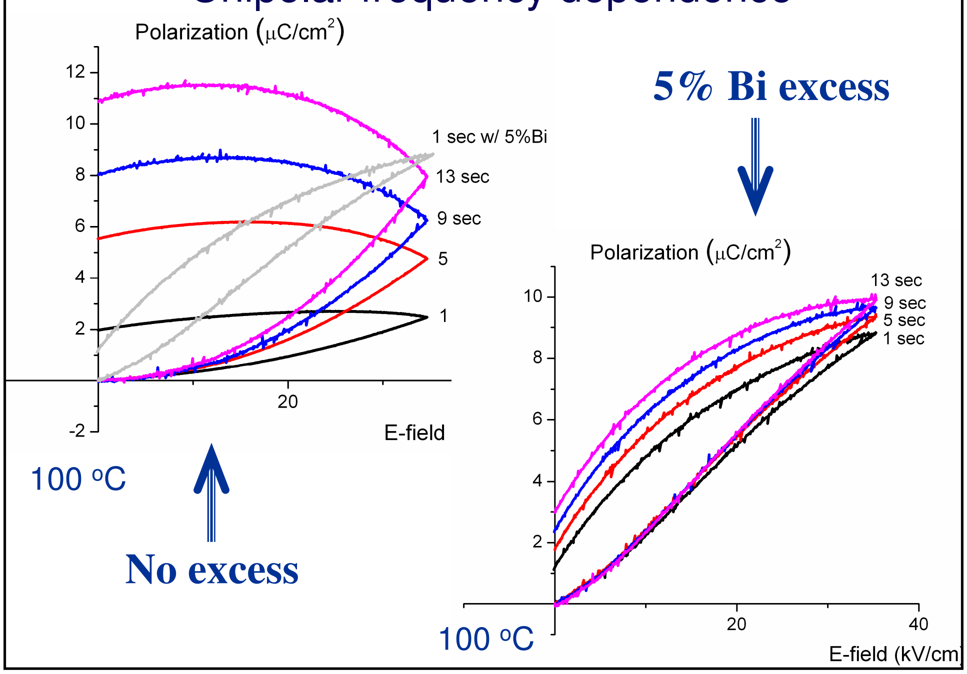
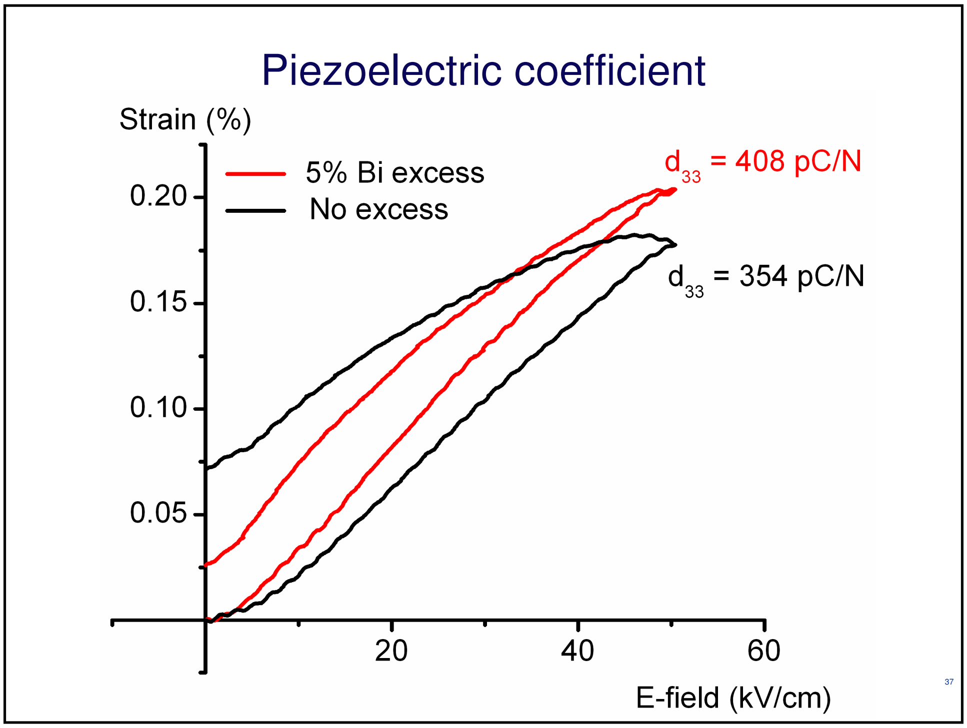
Ferroelectric Properties

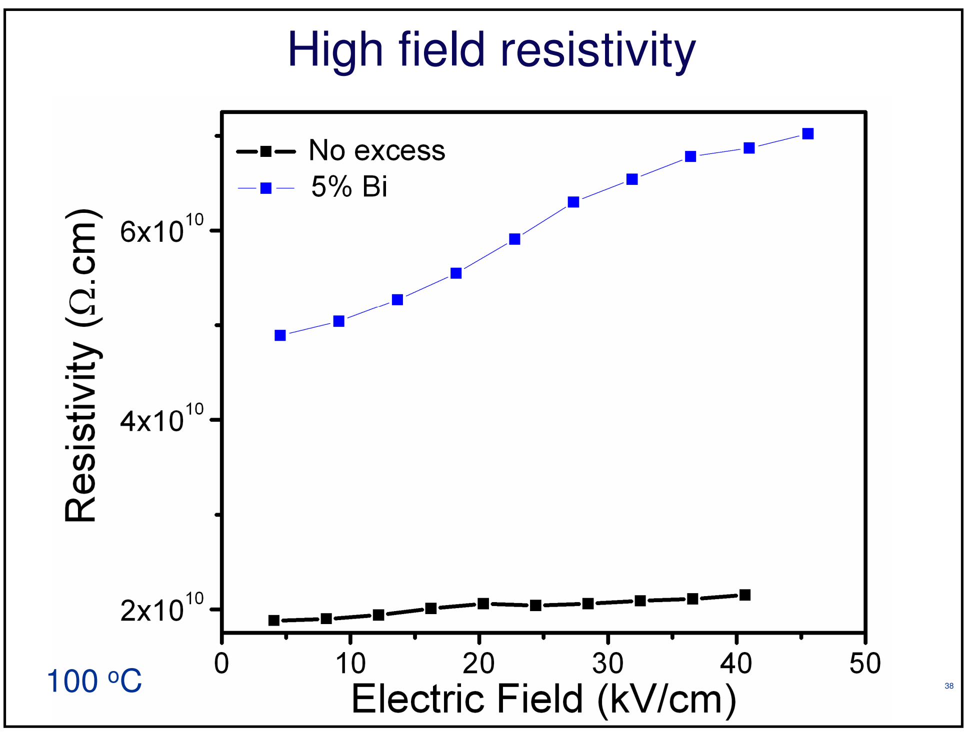


Unipolar frequency dependence



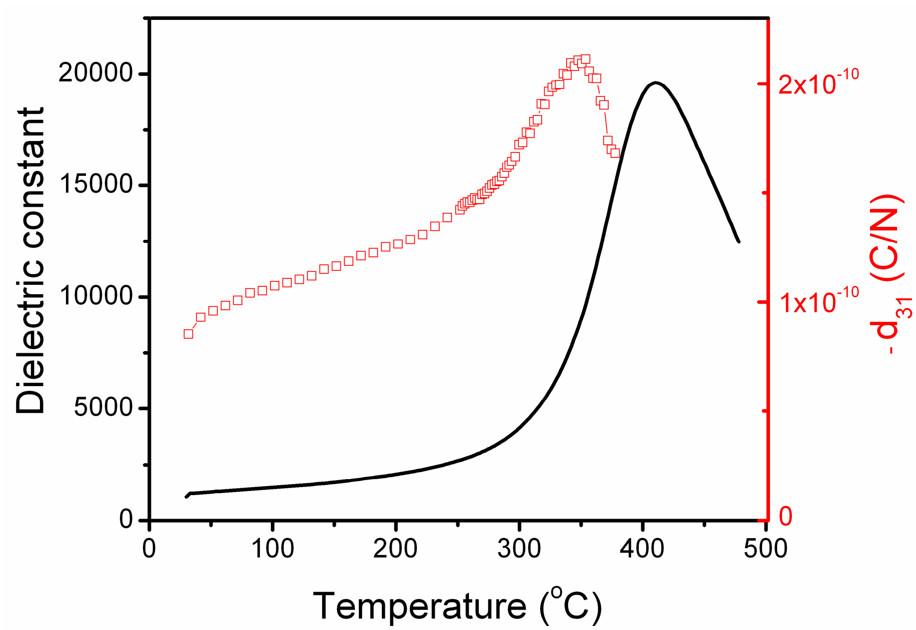


Piezoelectric coefficient
Strain (%)
5% Bi excess
No excess
d_{33} = 408 pC/N
d_{33} = 354 pC/N
0.20
0.15
0.10
0.05
20
40
60
E-field (kV/cm)
37



High field resistivity
No excess
5% Bi
Resistivity (Ω.cm)
6x10^10
4x10^10
2x10^10
0
10
20
30
40
50
Electric Field (kV/cm)
100 °C
38

Resonance-Antiresonance Measurements



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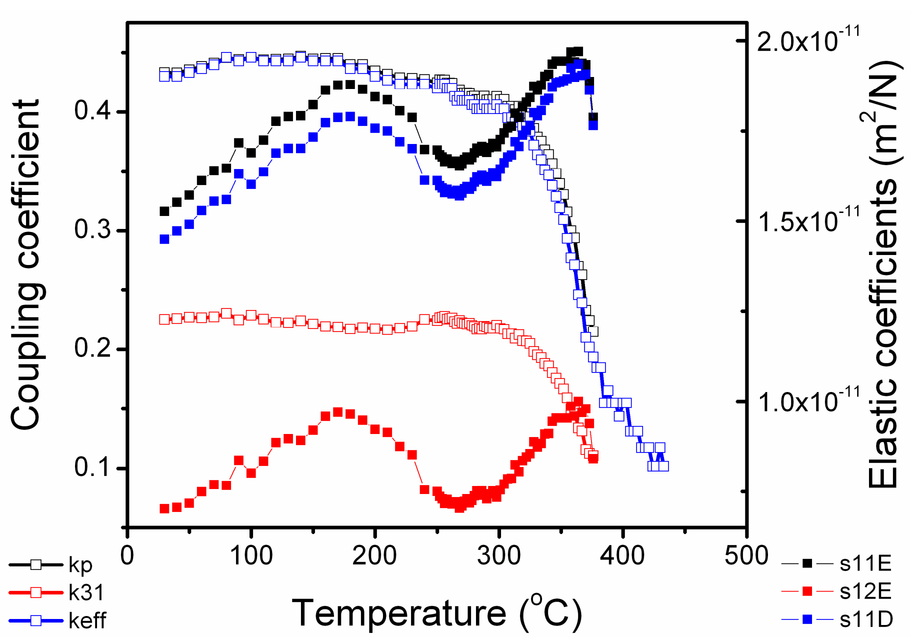
Resonance-Antiresonance Measurements



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Summary

- $BiScO_3$-$PbTiO_3$ ceramics with $T_C > 400^oC$ has been successfully processed.
- Despite the increase in T_C, excess Pb addition increases both the bulk conductivity and the grain boundary contribution to conductivity at elevated temperatures.
- Conductivity at elevated temperatures, that limits the operating temperature for actuators, has been greatly reduced by excess Bi additions.
- Excess Bi doping improves poling conditions resulting in enhanced piezoelectric coefficient (d_{33} = 408 pC/N).

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