Institut des Nanotechnologies de Lyon UMR 5270

Pyroelectricity

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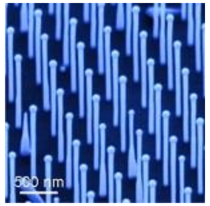
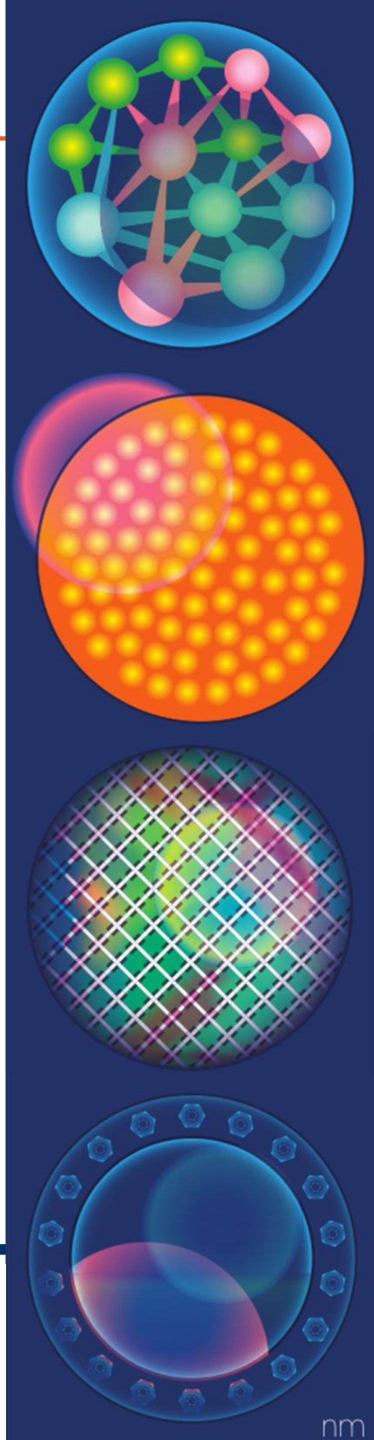


« EL NANO » thematic school, GDR NAME

June 11-16, 2023, Aussois



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Outline

Definition & general principles

Main applications

Pyroelectric materials

Pyroelectric measurements

Key results from literature

Conclusions

References

1. Definition & general principles

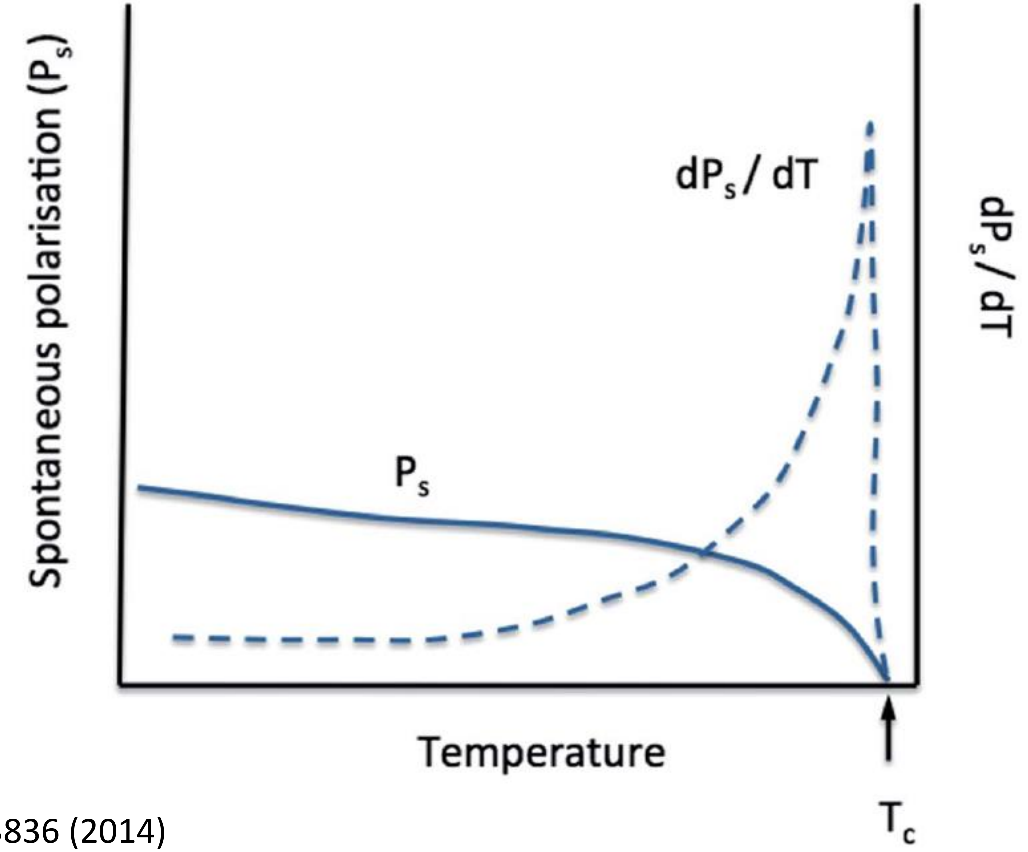
Definition & general principles

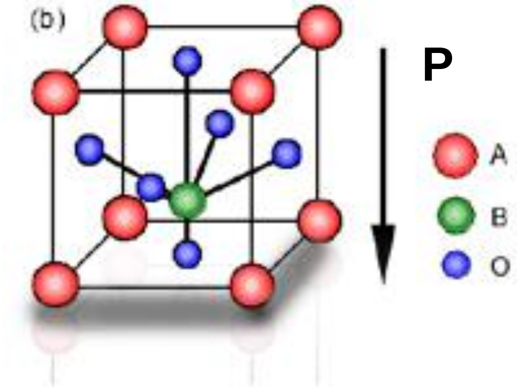
Pyroelectricity (PE): from the Greek word *pyr* meaning fire, and electricity

➔ **Variation of electric polarization P_s** (in a polar dielectric material) **with a change of temperature T**

$$p^{\sigma,\mathrm{E}} = \left(\frac{\mathrm{d}P_\mathrm{s}}{\mathrm{d}T}\right)_{\sigma,\mathrm{E}}$$

p: PE coeff. ($C.m^{-2}.K^{-1}$)

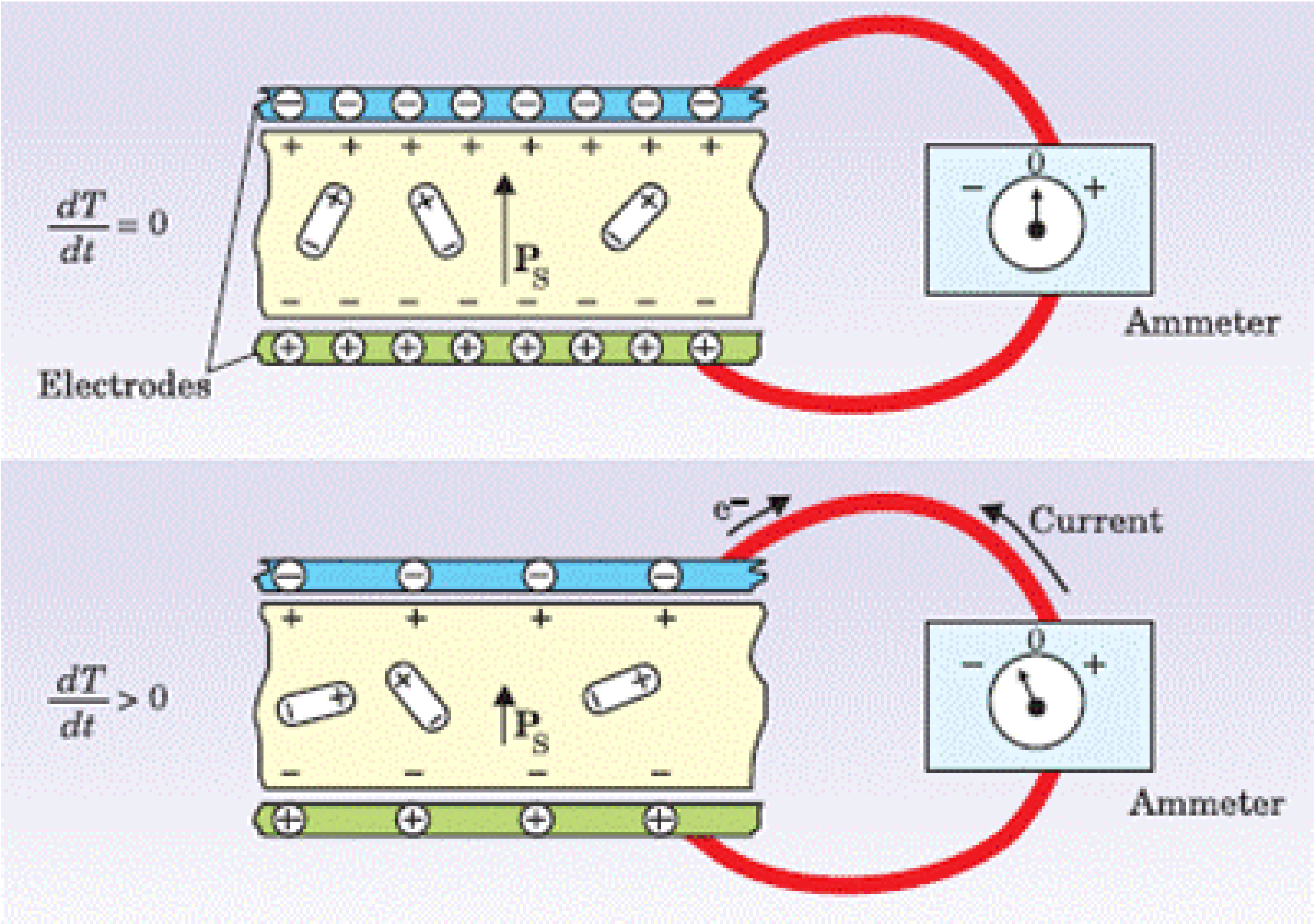


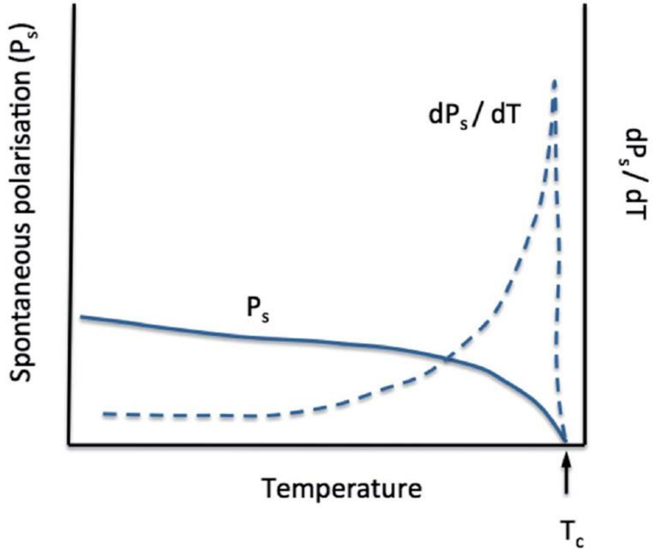


⇒ Enhancement can occur close to the phase transitions!

C.R. Bowen *et al.*, Energy Env. Sci. **7**, 3836 (2014)

Definition & general principles





Pyroelectric current i_p:

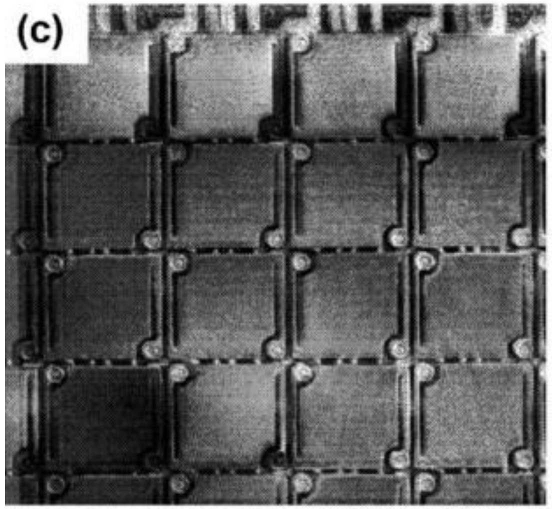
$$i_p = \frac{dQ}{dt} = pA\frac{dT}{dt}$$

S.B. Lang *et al*., Phys. Today **58**, 31 (2005)

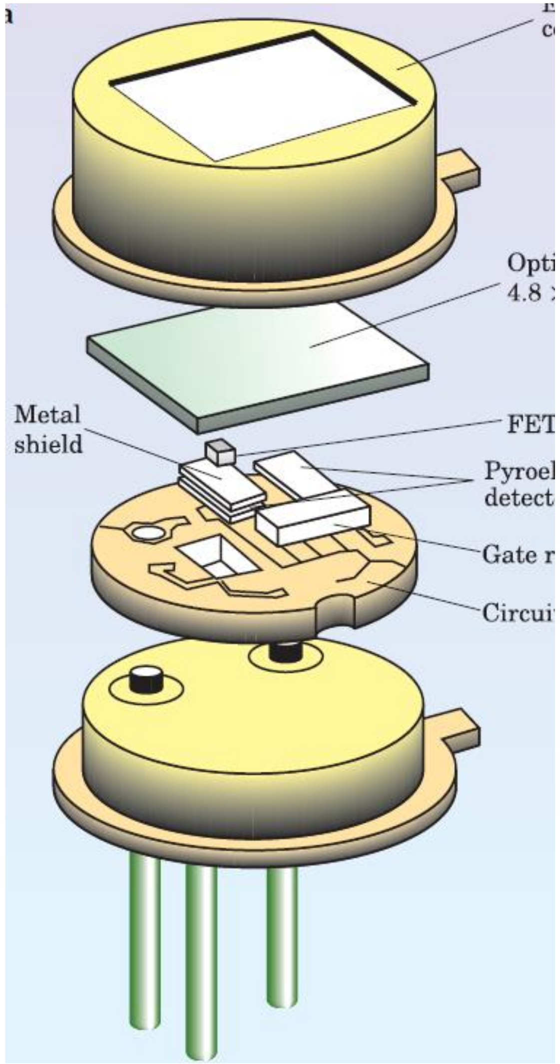
Main applications

IR sensors and imaging tools

Pyroelectricity: From Ancient Curiosity to Modern Imaging Tool



A 320 × 240 pyroelectric BST-based pixel array on an integrated circuit with 48.5 μm centers

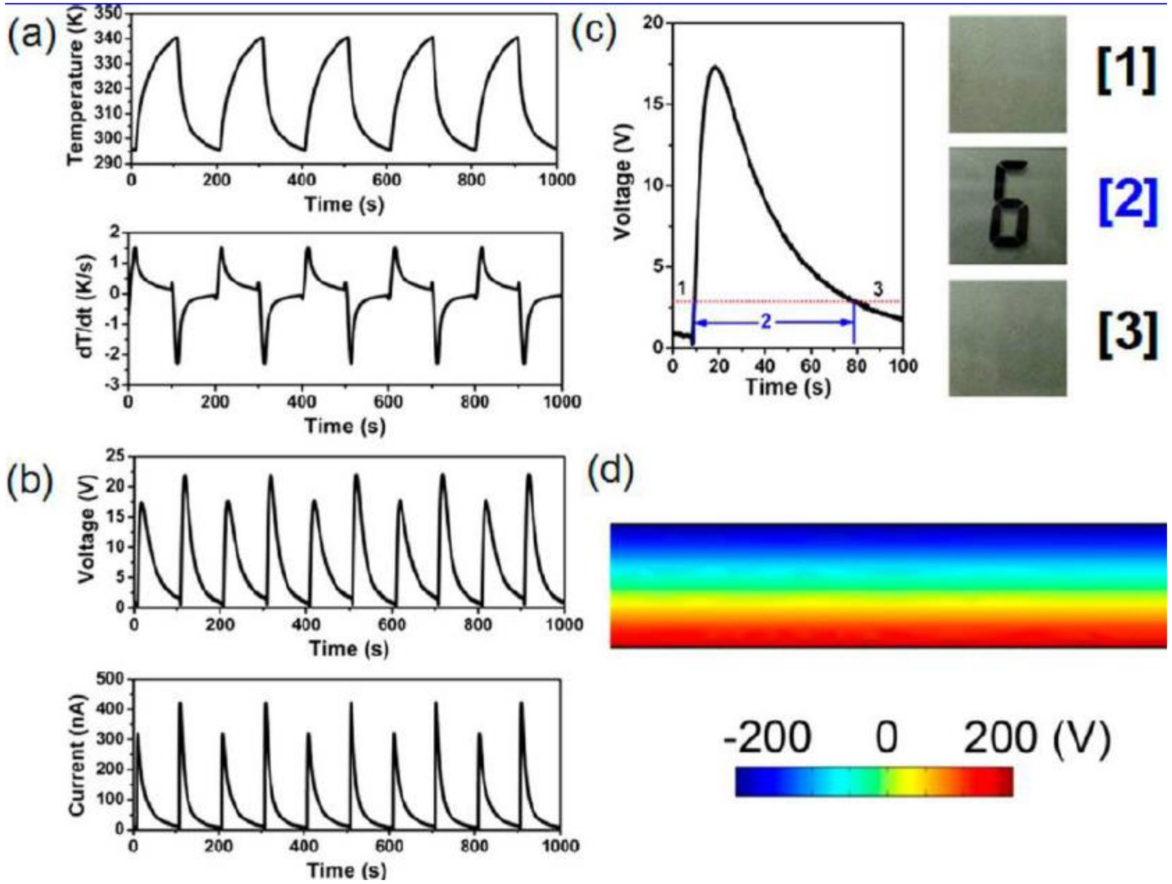


S.B. Lang *et al.*, Phys. Today **58**, 31 (2005); G. Velarde *et al.*, APL Mater. 9, 010702 (2021)

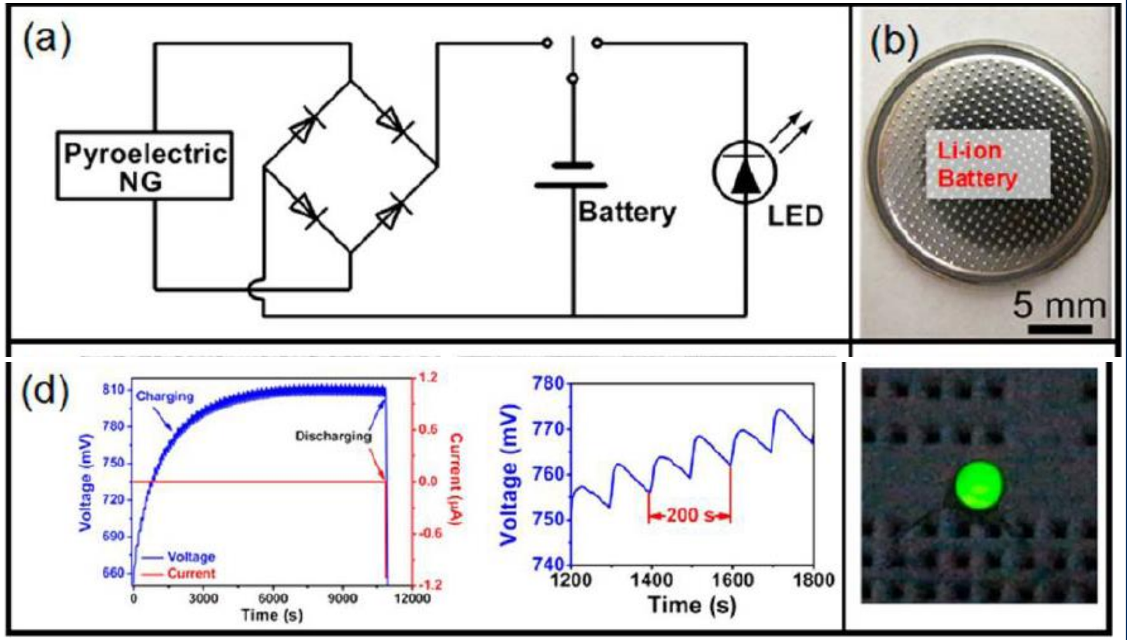
Main applications

Thermal energy harvesting

Pyroelectric Nanogenerators for Driving Wireless Sensors



From polycrystalline PZT (175 µm) thick film



Y. Yang et al., Nano Letters **12**, 6408 (2012)

Pyroelectric materials

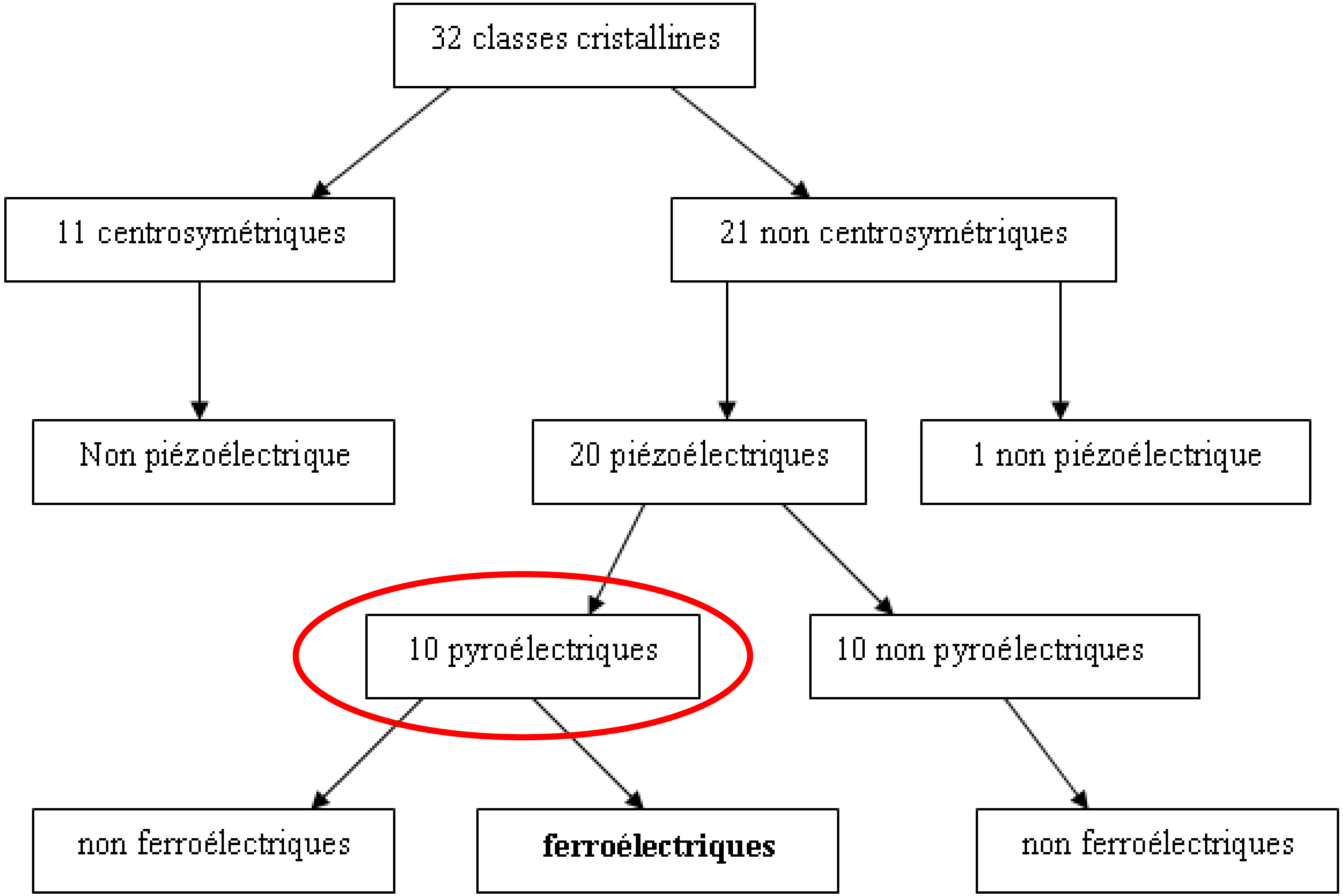
The best are ferroelectrics

Primary, secondary, and total pyroelectric coefficients of various materials. (Units are $\mu C/m^2 \cdot K$)

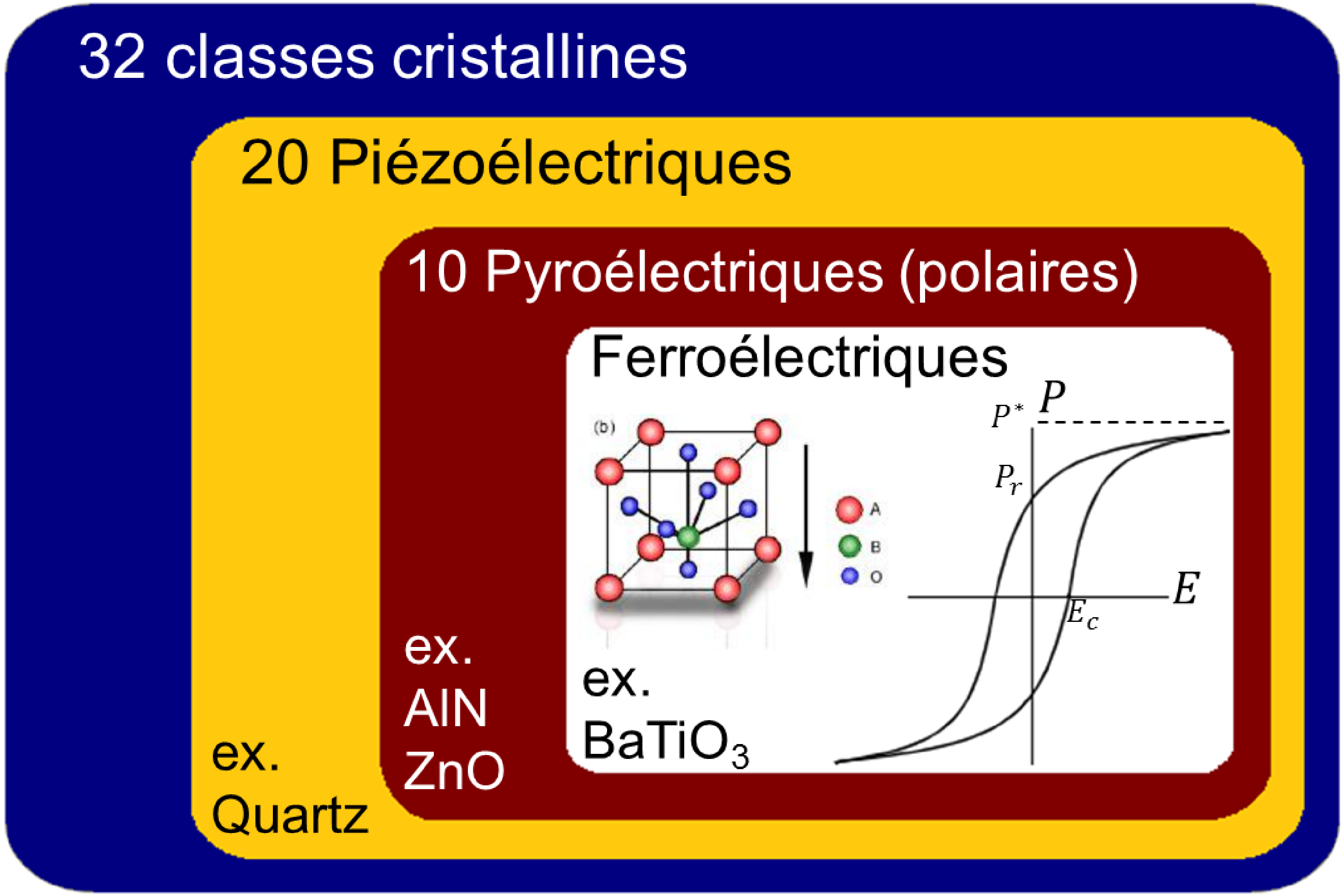
Material	Primary coefficient	Secondary coefficient	Total coefficient
Ferroelectrics			
Poled ceramic			
$BaTiO_3$	−260	+60	−200
$PbZr_{0.95}Ti_{0.05}O_3$	−305.7	+37.7	−268
Crystal			
$LiNbO_3$	−95.8	+12.8	−83
$LiTaO_3$	−175	−1	−176
$Pb_5Ge_3O_{11}$	−110.5	+15.5	−95
$Ba_2NaNb_5O_{15}$	−141.7	+41.7	−100
$Sr_{0.5}Ba_{0.5}Nb_2O_6$	−502	−48	−550
$(CH_2CF_2)_n$	−14	−13	−27
Triglycine sulfate	+60	−330	−270
Nonferroelectrics			
Crystal			
CdSe	−2.94	−0.56	−3.5
CdS	−3.0	−1.0	−4.0
ZnO	−6.9	−2.5	−9.4
Tourmaline	−0.48	−3.52	−4.0
$Li_2SO_4 \cdot 2H_2O$	+60.2	+26.1	+86.3

S.B. Lang *et al*., Phys. Today **58**, 31 (2005)

Pyroelectric materials



Definition & general principles

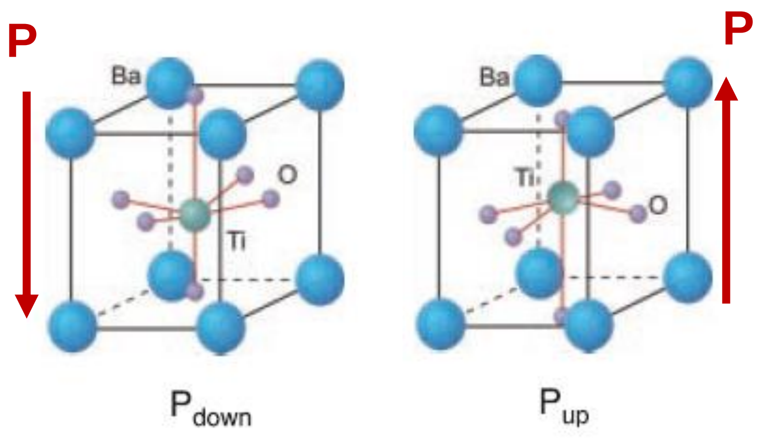


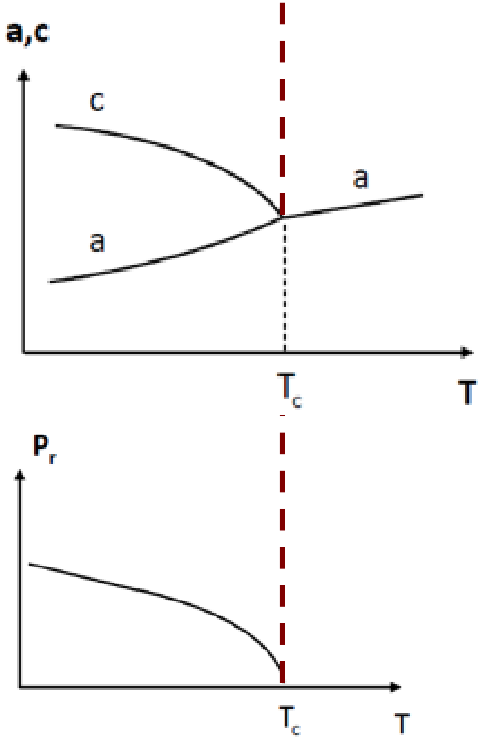
Ferroelectrics & phase changes accross Tc

Pb(Zr,Ti)O_3, PMN-PT,
(Ba,Sr)TiO_3,…

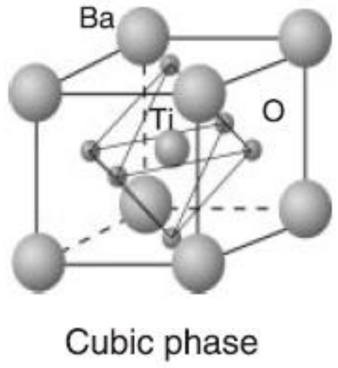
Tetragonal | **Cubic**

<u>Non-centrosymmetric</u>

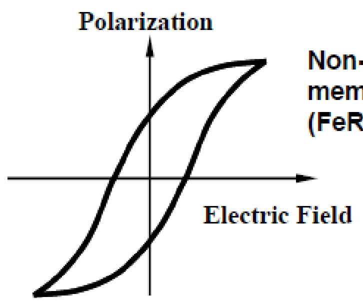




<u>Centrosymmetric</u>

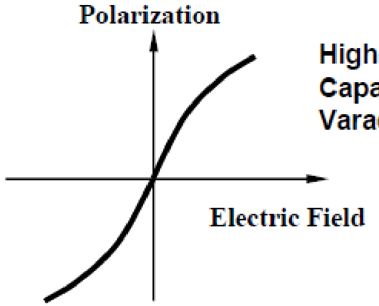


Ferroelectric $T < T_c$



Non-volatile memories (FeRAM)

Paraelectric $T > T_c$

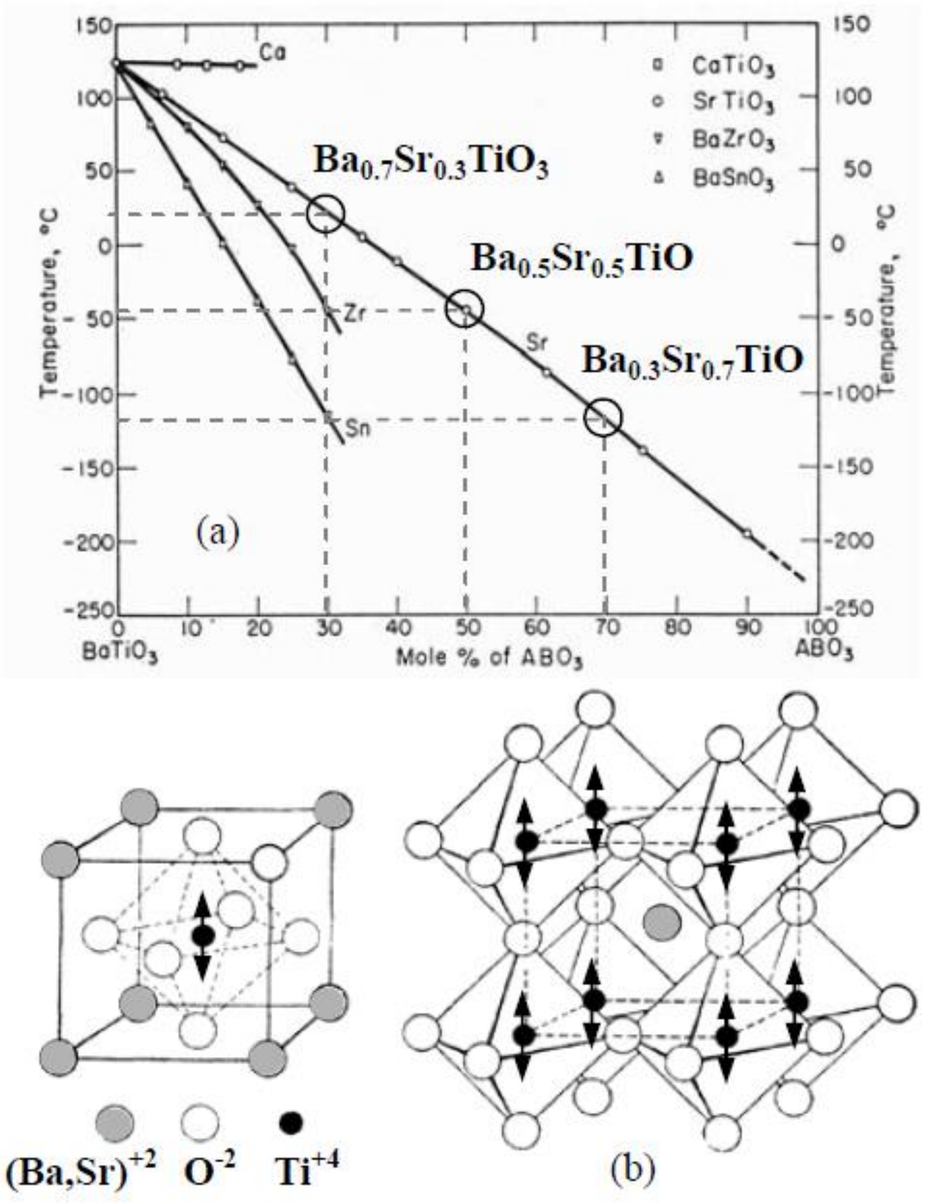


High-κ Capacitors & Varactors

Tunable Curie temperature (T_c)

by cationic substitution

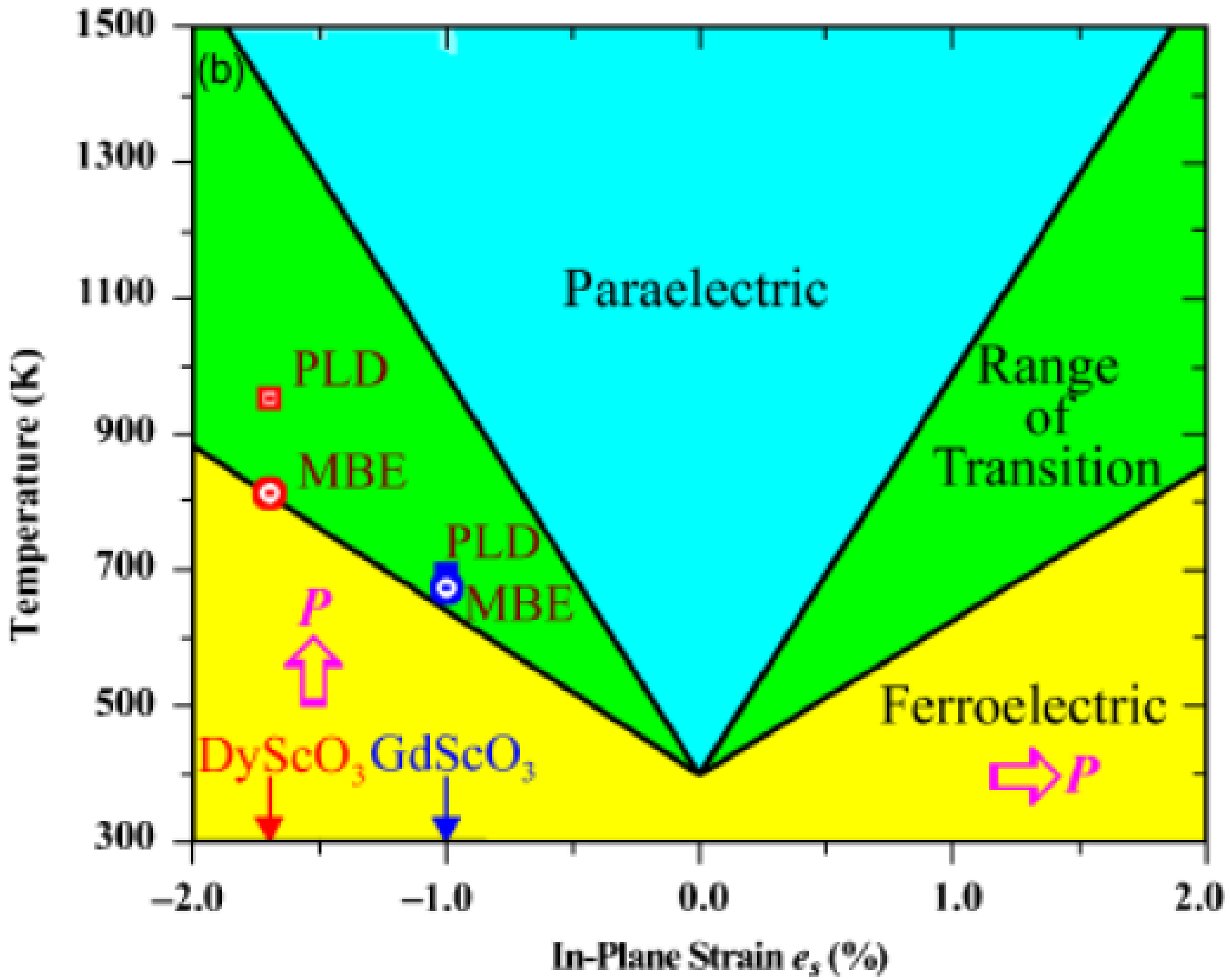
Exemple: (Ba,Sr)TiO_3 (ferroelectric)



Tunable Curie temperature (T_c)

by epitaxial strain

Exemple: $BaTiO_3$ (ferroelectric at RT)



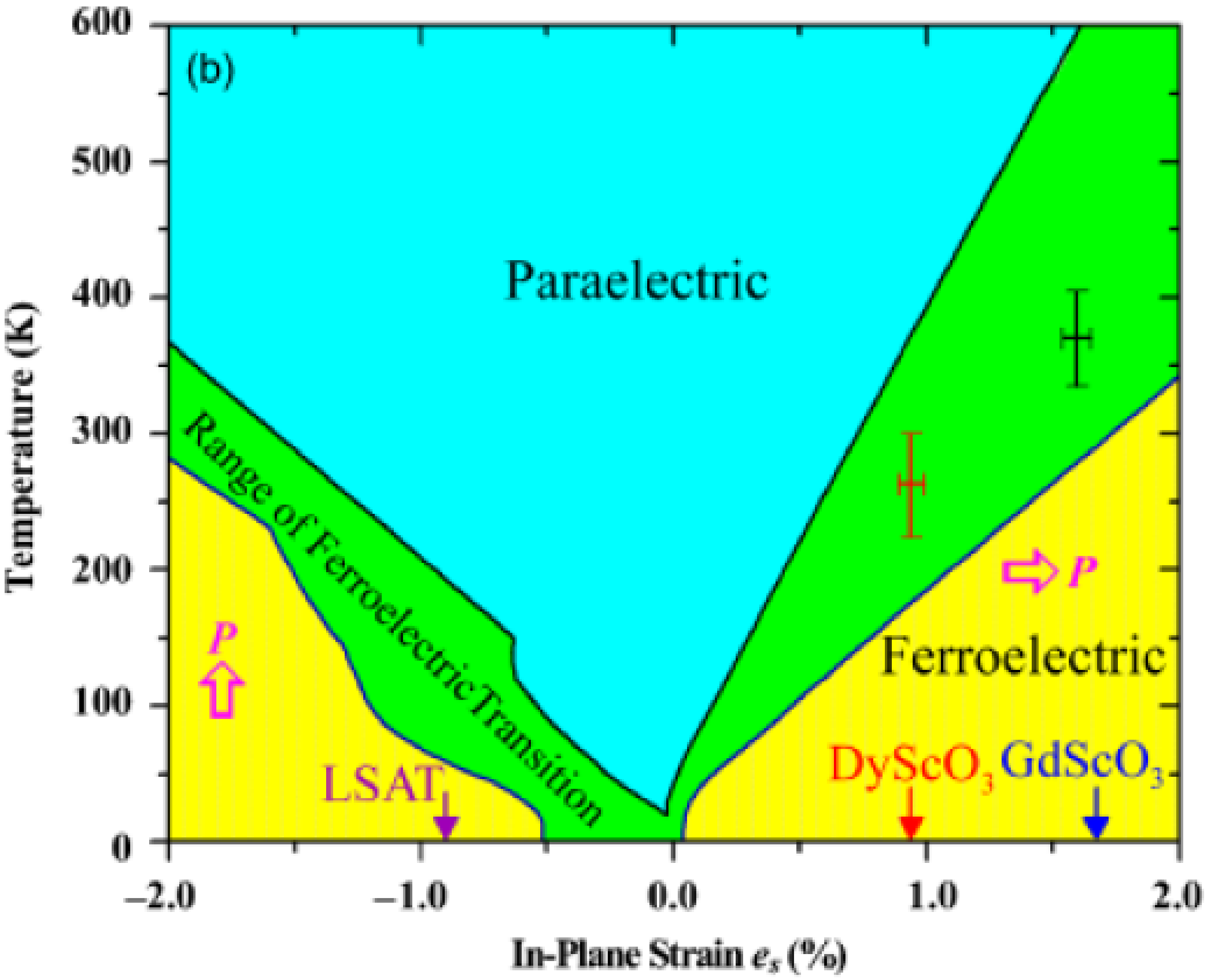
K.J. Choi *et al.*, Science 306, 1005 (2004)

Tunable Curie temperature (T_c)

by epitaxial strain

Exemple: $SrTiO_3$ (bulk dielectric at RT)

➔ Become ferroelectric when strained!



J.H Haeni *et al.*, Nature 430, 758 (2004)

Y.L. Li et al., PRB 73, 184112 (2006)

Pyroelectric figure of merit & estimation of harvested energy

For thermal energy harvesting

$$\text{FOM} = \frac{p^2}{\varepsilon_{33}^T}.$$

Harvested energy:

$$W_D = \frac{p^2}{\varepsilon_{33}} . \Delta T^2$$

Various slightly different FoM exist

k^2: electro-thermal coupling factor

p (μC m^{-2} K^{-1})	$\varepsilon_{33}/\varepsilon_0$	c_E (MJ m^{-3} K^{-1})	T_C (°C)	k_{therm} (W m^{-1} K^{-1})	k^2 (T_{hot} = 300 K) $k^2 = \frac{p^2 T_{hot}}{c_E \varepsilon_{33}^\sigma}$	F_i (×10^{-10}) (m V^{-1}) $F_i = \frac{p}{c_E}$	F_v (m^2 C^{-1}) $F_v = \frac{p}{c_E \varepsilon_{33}^\sigma}$	F_E (×10^{-11}) (J m^{-3} K^{-2}) $F_E = \frac{p^2}{\varepsilon_{33}^\sigma}$	F'_E (×10^{-11}) (m^3 J^{-1}) $F'_E = \frac{p^2}{\varepsilon_{33}^\sigma (c_E)^2}$

C.R. Bowen *et al.*, Energy Env. Sci. (2014); & G. Sebald *et al.*,…

Pyroelectric materials

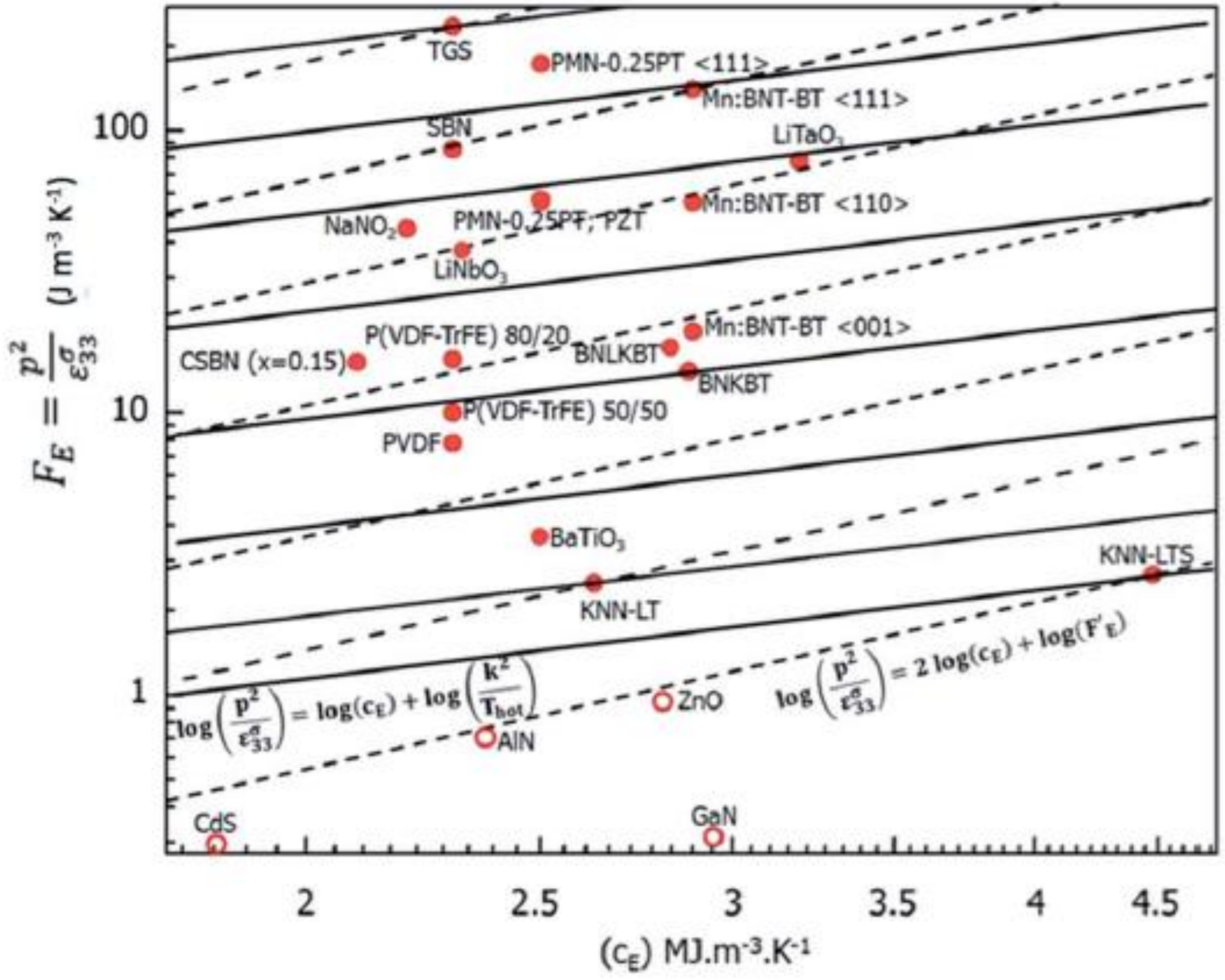
Material	Ferroelectric	p (μC m^{-2} K^{-1})	$\varepsilon_{33}/\varepsilon_0$	c_E (MJ m^{-3} K^{-1})	T_C (°C)	k_{therm} (W m^{-1} K^{-1})	k^2 (T_{hot} = 300 K) $k^2 = \frac{p^2 T_{hot}}{c_E \varepsilon^{\sigma}_{33}}$	F_i ($\times 10^{-10}$) (m V^{-1}) $F_i = \frac{p}{c_E}$	F_v (m^2 C^{-1}) $F_v = \frac{p}{c_E \varepsilon^{\sigma}_{33}}$	F_E ($\times 10^{-11}$) (J m^{-3} K^{-2}) $F_E = \frac{p^2}{\varepsilon^{\sigma}_{33}}$	F'_E ($\times 10^{-11}$) (m^3 J^{-1}) $F'_E = \frac{p^2}{\varepsilon^{\sigma}_{33}(c_E)^2}$	Reference
Triglycine sulphide SC (TGS)	✓	−280 (60, −330)	38	2.3	49	0.65	0.030408	1.21	0.362	233.13	4.41	14 and 18
Lead magnesium niobate-lead titanate PMN-0.25PT ⟨111⟩ (SC)	✓	−1790	2100	2.5	121	2.5 (PMN-0.34PT)	0.020688	7.16	0.039	172.40	2.76	53
Mn : BNT-BT ⟨111⟩ (SC)	✓	−588	279	2.89	—	—	0.014536	2.03	0.082	140.03	1.68	73
Strontium barium niobate (SBN x = 0.5)	✓	−550	400	2.3	125	0.6	0.011146	2.39	0.068	85.45	1.62	14 and 138
Lithium tantalate ($LiTaO_3$)	✓	−176 (−175, −1)	47	3.2	665	3.9	0.006982	0.55	0.132	74.47	0.73	11 and 18
PMN-0.25PT	✓	−746	2100	2.5			0.003593	2.98	0.031	29.94	0.48	31
Lead zirconate titanate (PZT)	✓	−380	290	2.5	200	0.8	0.006752	1.52	0.059	56.26	0.90	14 and 139
Sodium nitride ($NaNO_2$)	✓	−40	4	2.2	164	2.2	0.006163	0.182	0.514	45.20	0.93	14
Mn : BNT-BT ⟨110⟩ (SC)	✓	−513	535	2.89	—	—	0.005770	1.77	0.037	55.58	0.67	73
CSBN x = 0.15	✓	−361	972	2.1	—	—	0.002164	1.71	0.020	15.15	0.34	140
Mn : BNT-BT ⟨001⟩ (SC)	✓	−380	835	2.89	—	—	0.002028	1.31	0.018	19.54	0.23	73
P(VDF-TrFE) 80/20	✓	−31	7	2.3	135	0.14	0.002023	0.13	0.218	15.51	0.29	14
BNLKBT	✓	−360	858	2.83	—	—	0.001809	1.27	0.017	17.07	0.21	141
BNKBT	✓	−325	853	2.88	—	—	0.001457	1.12	0.015	13.99	0.17	141
P(VDF-TrFE) 50/50	✓	−40	18	2.3	49	0.14	0.001310	0.17	0.109	10.04	0.19	14
PVDF	✓	−27 (−14, −13)	9	2.3	80	0.14	0.001194	0.10	0.147	9.15	0.17	11, 12 and 14
KNN-LT	✓	−165	1230	2.63	—	—	0.000285	0.62	0.006	2.50	0.04	141
KNN-LTS	✓	−190	1520	4.48	—	—	0.000180	0.42	0.003	2.68	0.01	141
Lithium niobate ($LiNbO_3$)	✓	−83 (−95.9, +12.9)	28.7	2.32	1210	1.1	0.003507	0.35	0.141	27.12	0.50	12, 39, 77, 138, 142 and 143
Barium titanate ($BaTiO_3$)	✓	−200 (−260, +60)	1200	2.5	120	3.0	0.000452	0.80	0.008	3.77	0.06	12, 139, 142 and 144
Zinc oxide (ZnO)	✗	−9.4 (−6.9, −2.5)	11	2.8	—	147	0.000097	0.034	0.034	0.91	0.01	18, 142, 145 and 146
Aluminium nitride (AlN)	✗	6–8	10	2.38	—	140	0.000070	0.033	0.038	0.55	0.01	142 and 147
Cadmium sulphide (CdS)	✗	−4 (−3, −1)	10.3	1.82	—	40	0.000029	0.022	0.024	0.18	0.005	12, 142, 148 and 149
Gallium nitride (GaN)	✗	−4.8	11	2.97	—	150	0.000024	0.016	0.017	0.24	0.003	142, 150 and 151

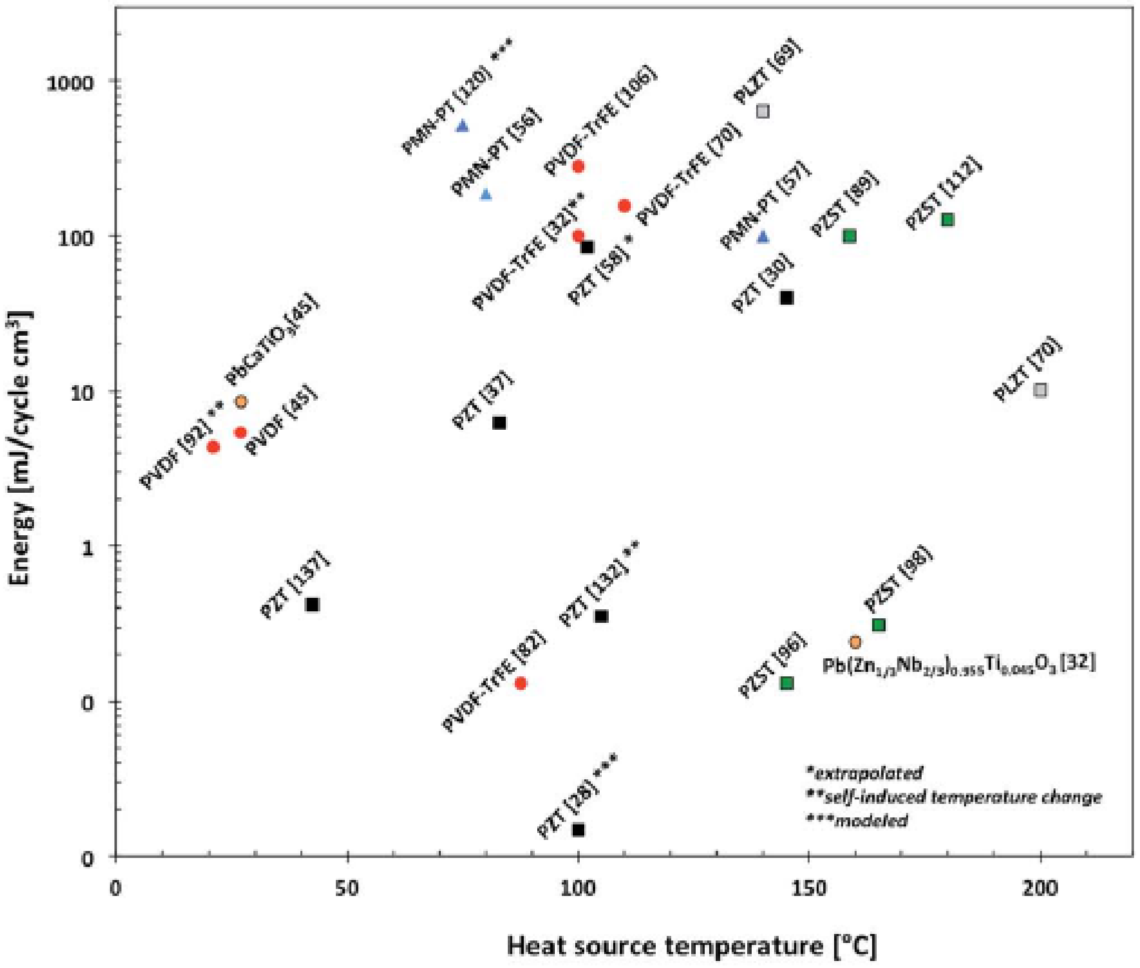
C.R. Bowen *et al.*, Energy Env. Sci. (2014)

Pyroelectric materials

The main bests contain Pb!... (PMN-PT, PZT,…)

PMN-PT = $Pb(Mg_{1/3}Nb_{2/3})O_3–PbTiO_3$





C.R. Bowen *et al.*, Energy Env. Sci. (2014)

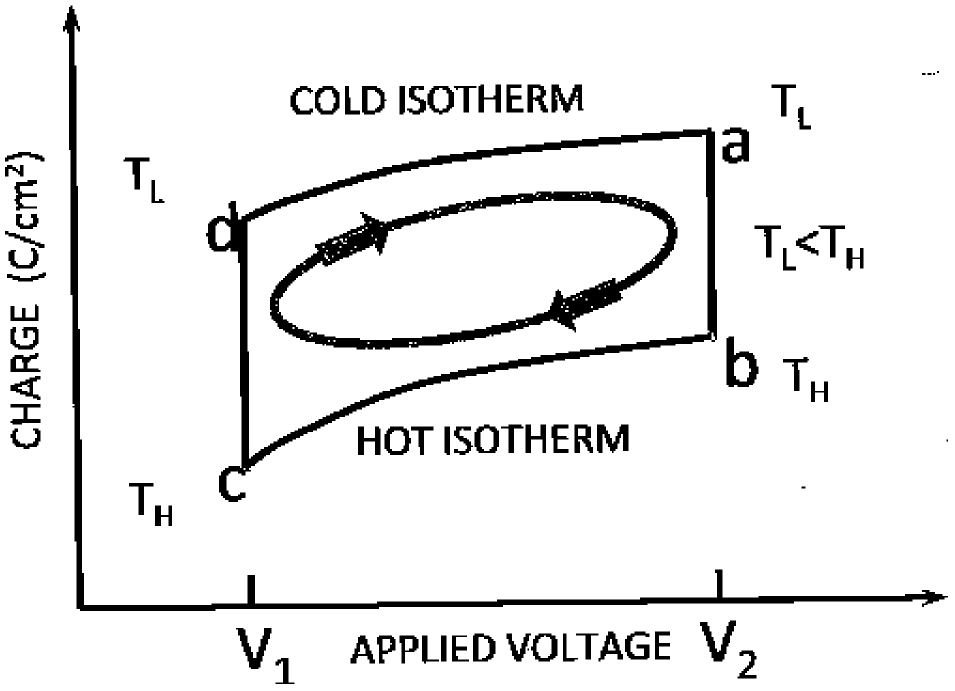
➔ Need to find Pb-free pyroelectrics!...

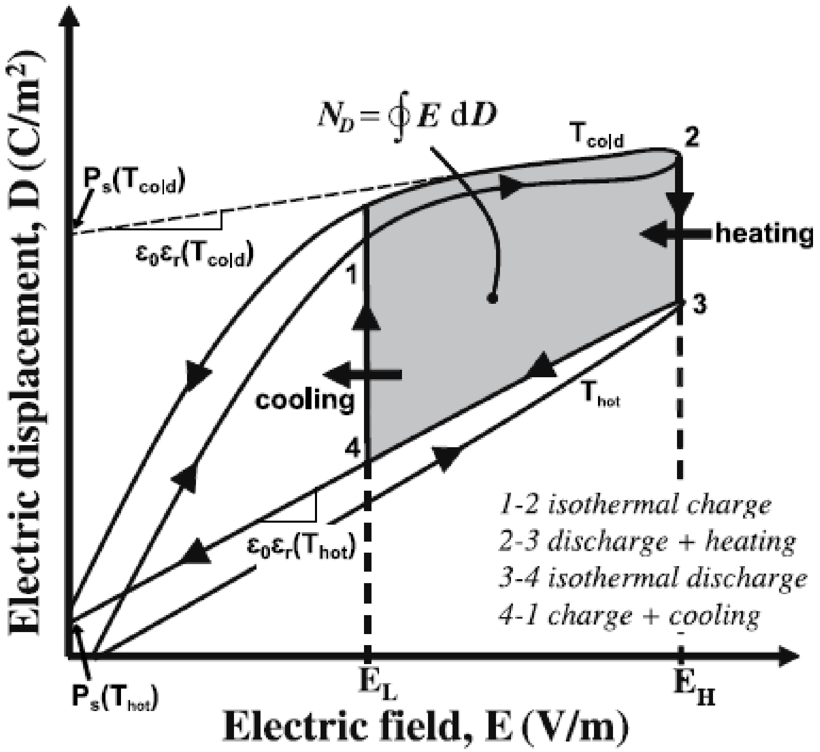
Cycling for thermal energy harvesting

Thermal energy harvesting by thermodynamical cycles

Carnot, Olsen, Ericson,…

$$W_D = \frac{p^2}{\varepsilon_{33}} . \Delta T^2$$



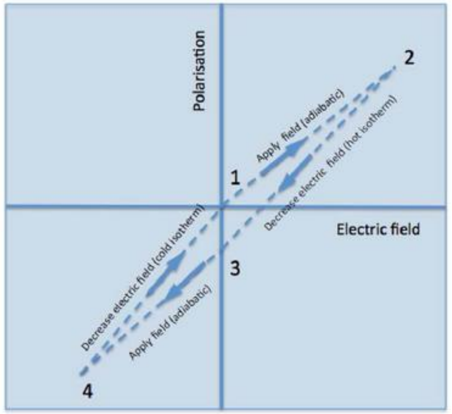


C.R. Bowen *et al.*, Energy Env. Sci. (2014)

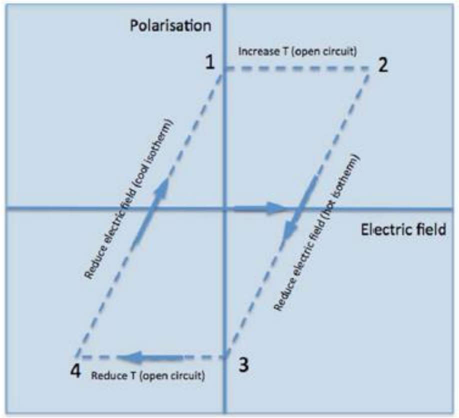
Cycling for thermal energy harvesting

Optimized cycles: synchronized (SECE, SSHI, SSDI,…) originally developed for piezoelectrics

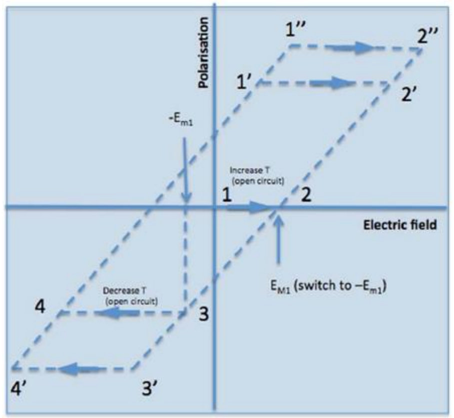
Carnot



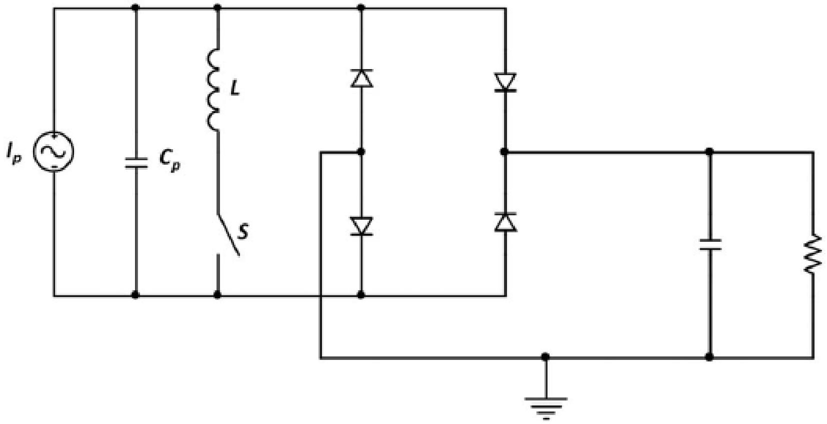
SECE



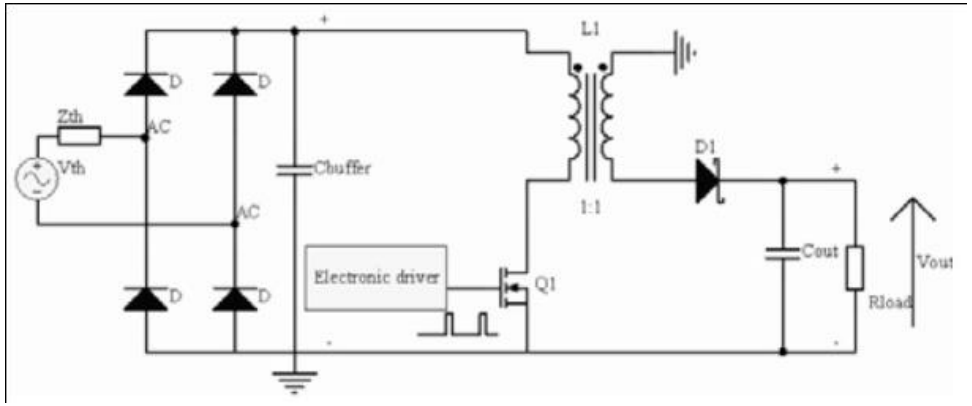
SSDI



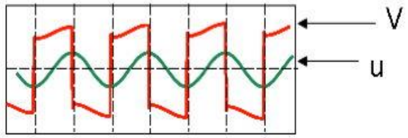
Optimized circuitries…

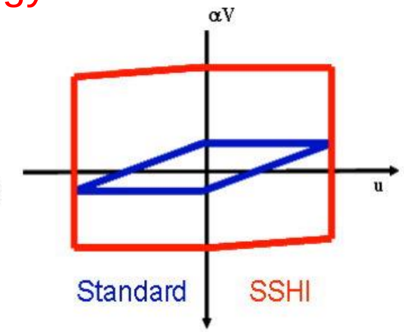


AC/DC converter & DC/DC converter to adapt the impedance



SSHI: enhanced energy exctraction (2-10 times)!





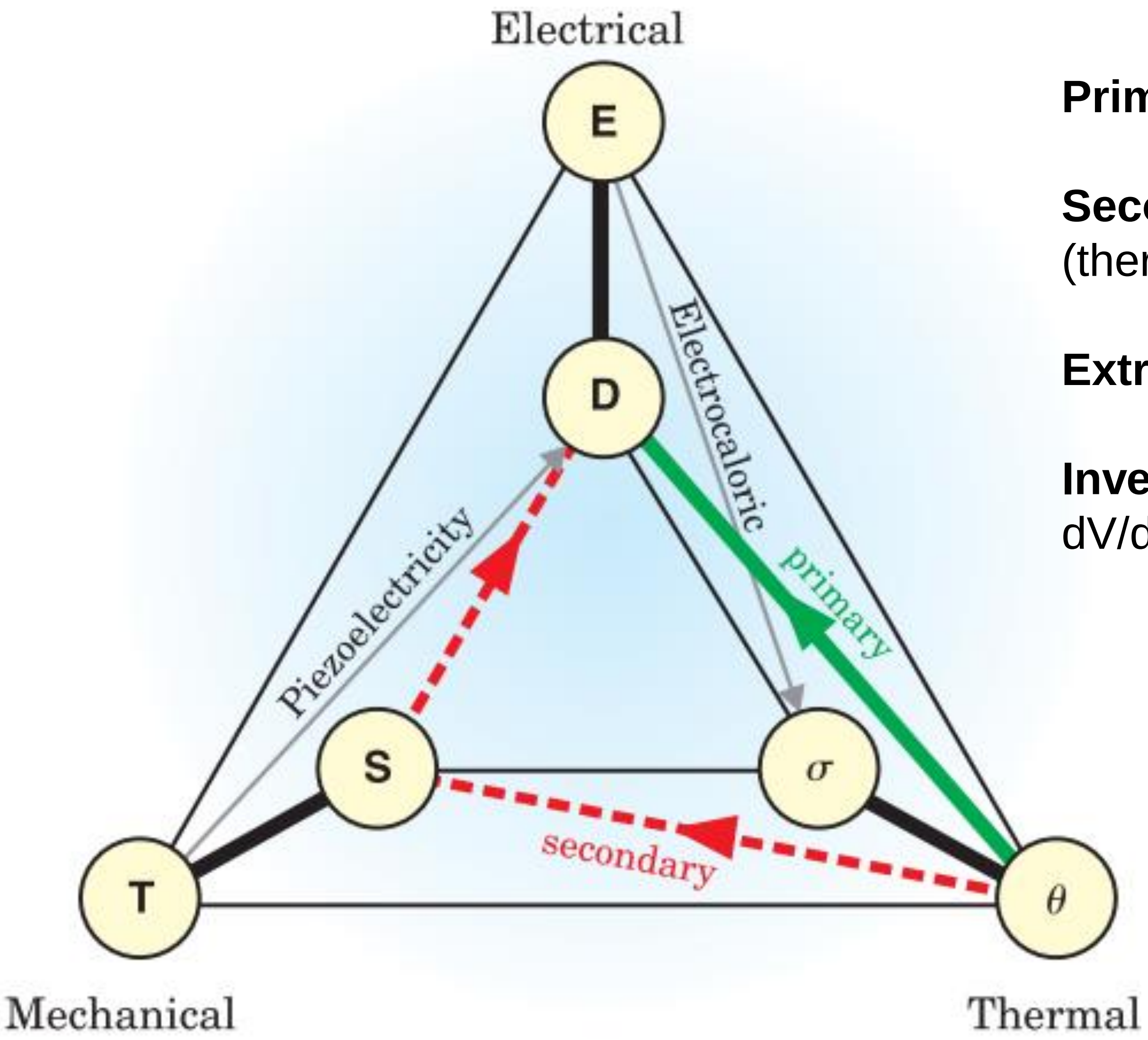
C.R. Bowen *et al.*, Energy Env. Sci. (2014); & G. Sebald *et al.*, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **(2008)**; also M. Lallart *et al.*,… (LGEF)

The different pyroelectric contributions

&

couplings

Heckmann diagram



Primary: purely thermal effect on electric polar

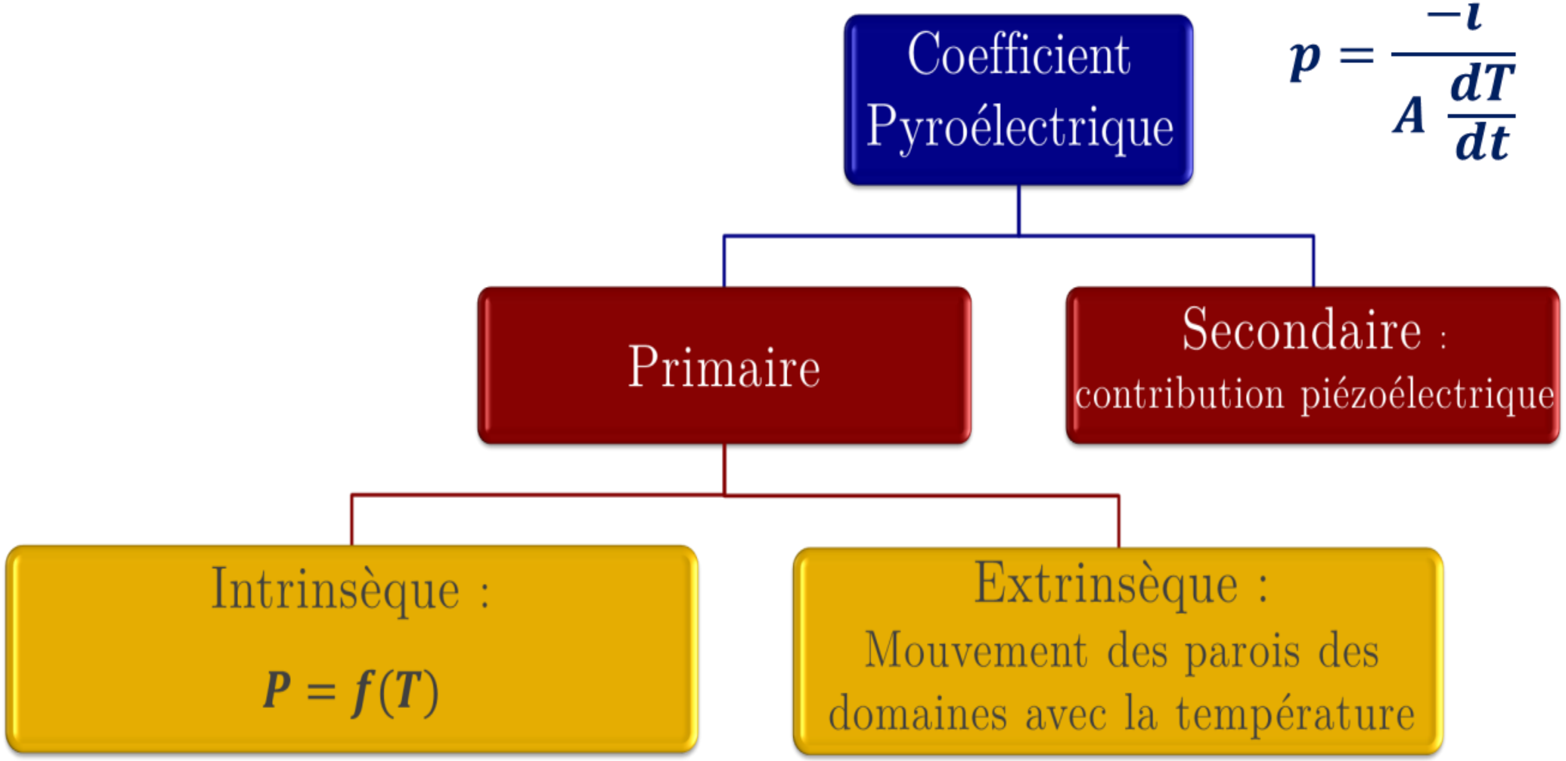
Secondary: linked with strain
(thermal expansion & piezoelectricity)

Extrinsic: dynamic effect due to domain wall motion

Inverse effect: Electrocaloric (EC)
$dV/dt \Rightarrow \Delta T$

S.B. Lang *et al*., Phys. Today **58**, 31 (2005)

Pyroelectric contributions



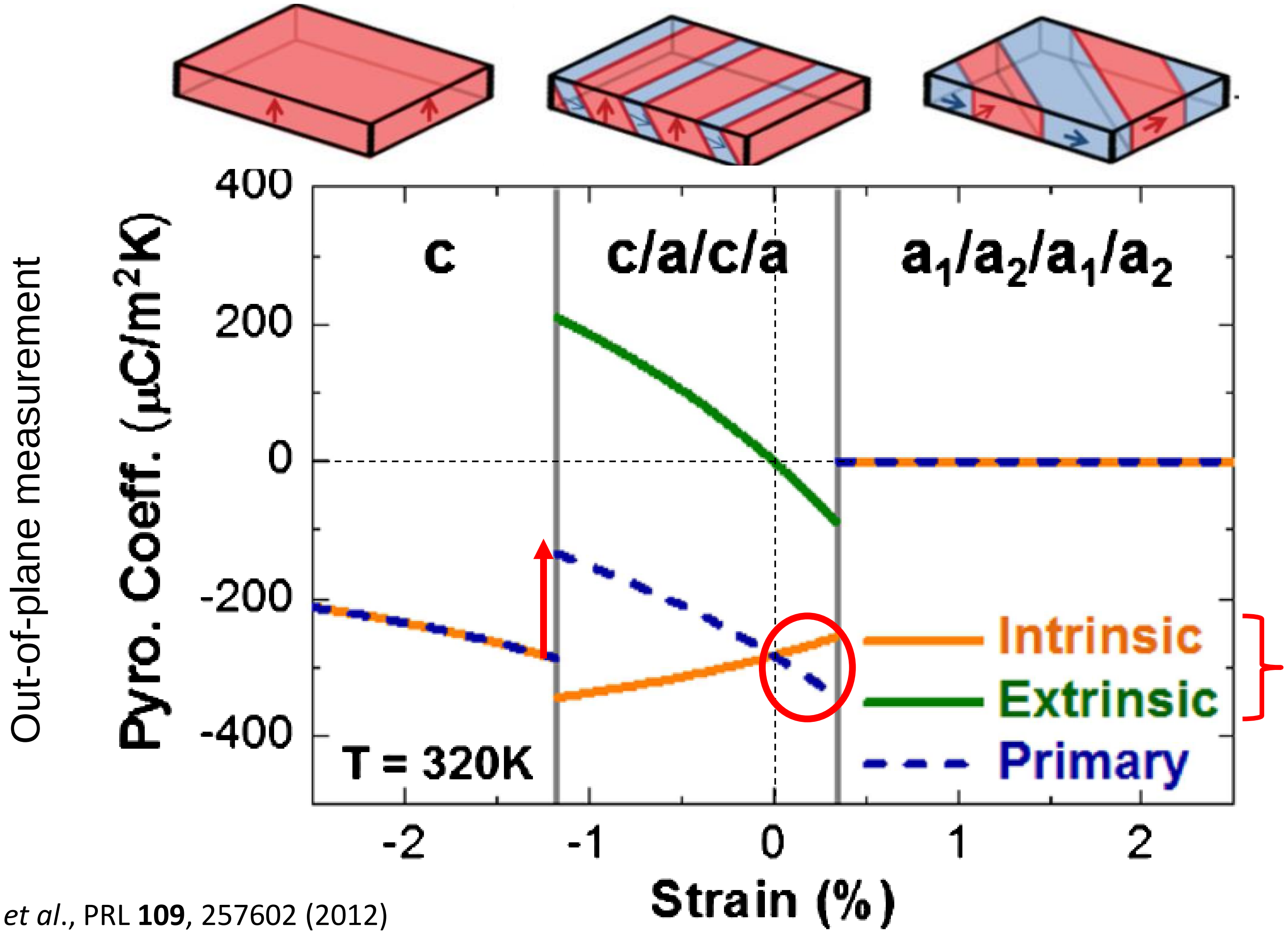
$$p = \frac{-i}{A\,\frac{dT}{dt}}$$

$$p = dP/dT$$

R. Moalla, PhD thesis, INL-ECL (2016)

Pyroelectric contributions

Different effects: function of FE domain orientations



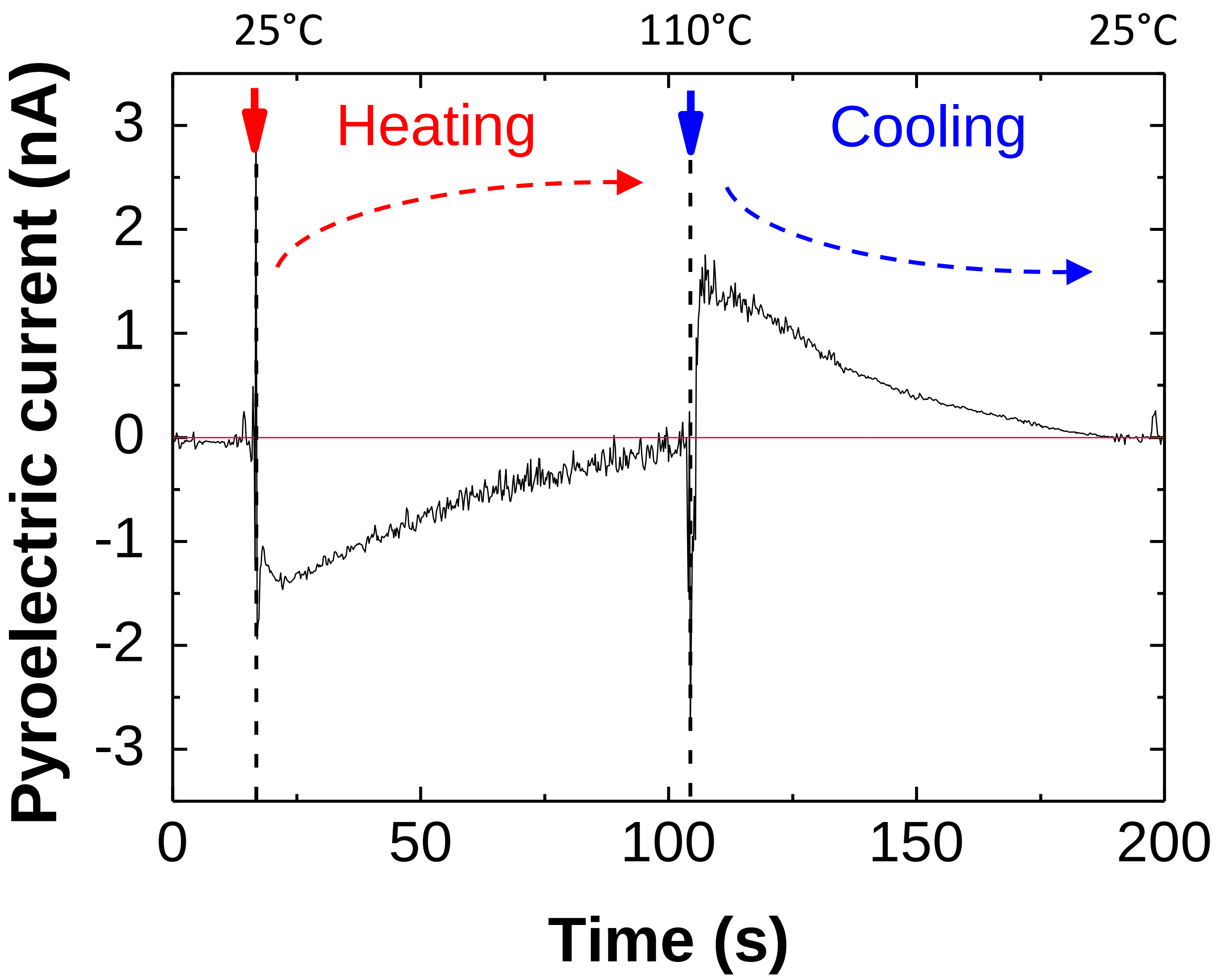
J. Karthik *et al.*, PRL **109**, 257602 (2012)

Pyroelectric measurements

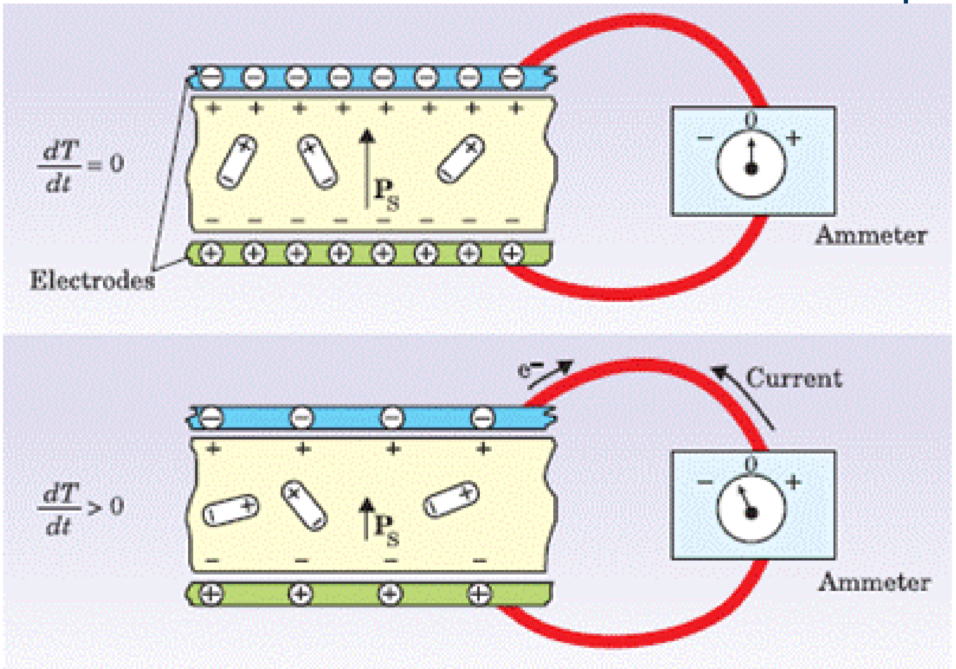
Pyroelectric effect: measurements

Direct measurements (all the pyroelectric contributions)

@INL



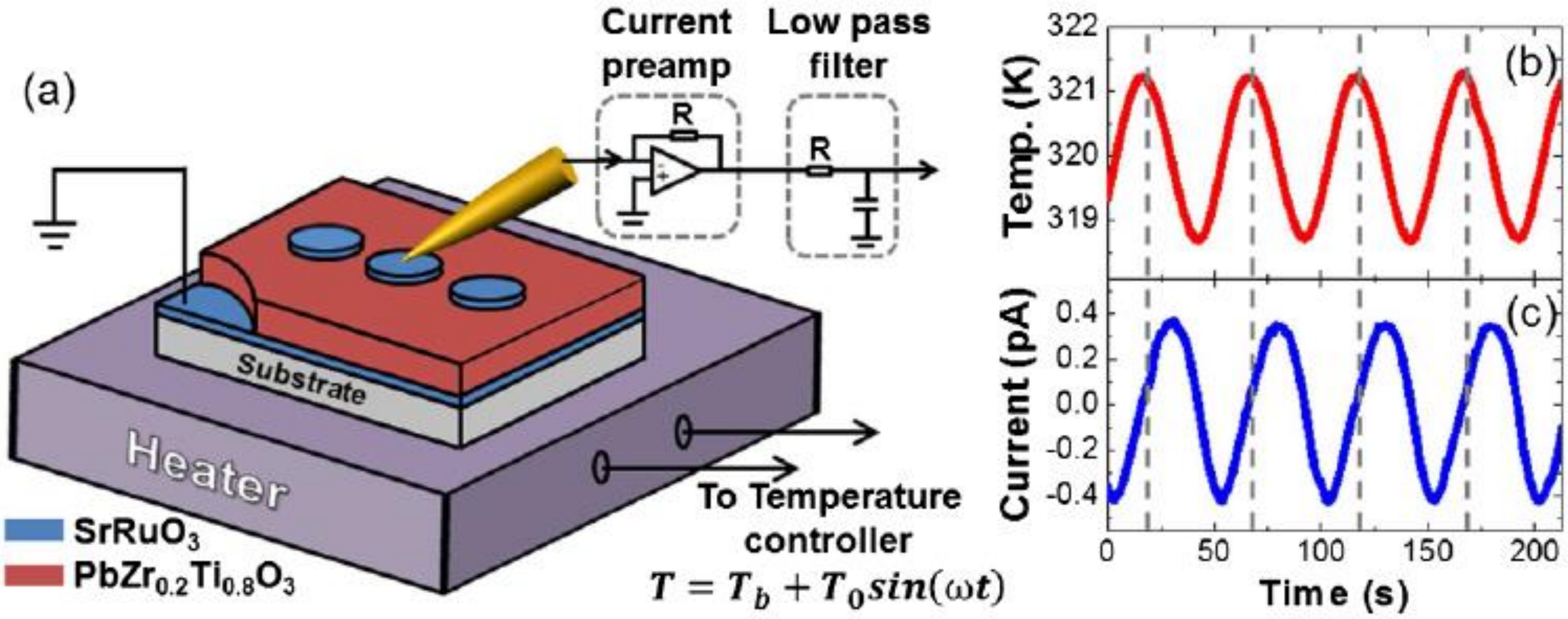
MIM capacitor



R. Moalla *et al.*, Thin Sol. Films **601**, 80 (2016)

Pyroelectric effect: measurements

Direct measurements (all the pyroelectric contributions)

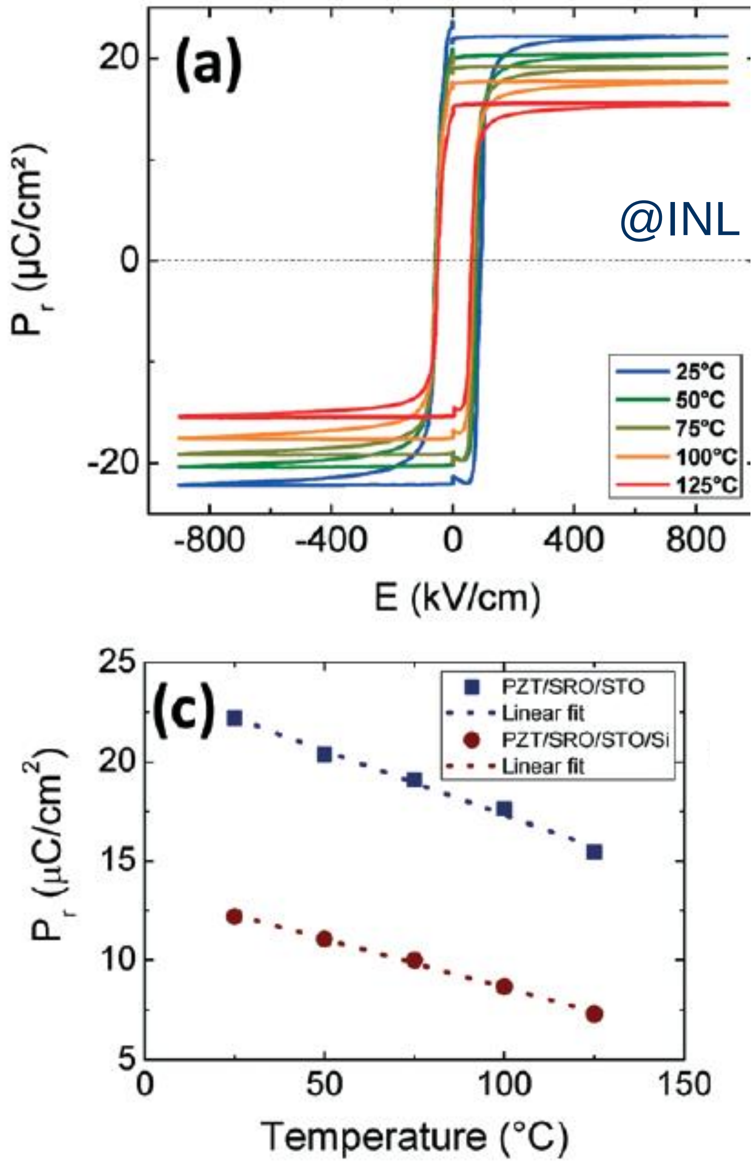


J. Karthik et al., PRL **109**, 257602 (2012)

Pyroelectric effect: measurements

Indirect measurements

(no extrinsic effect)



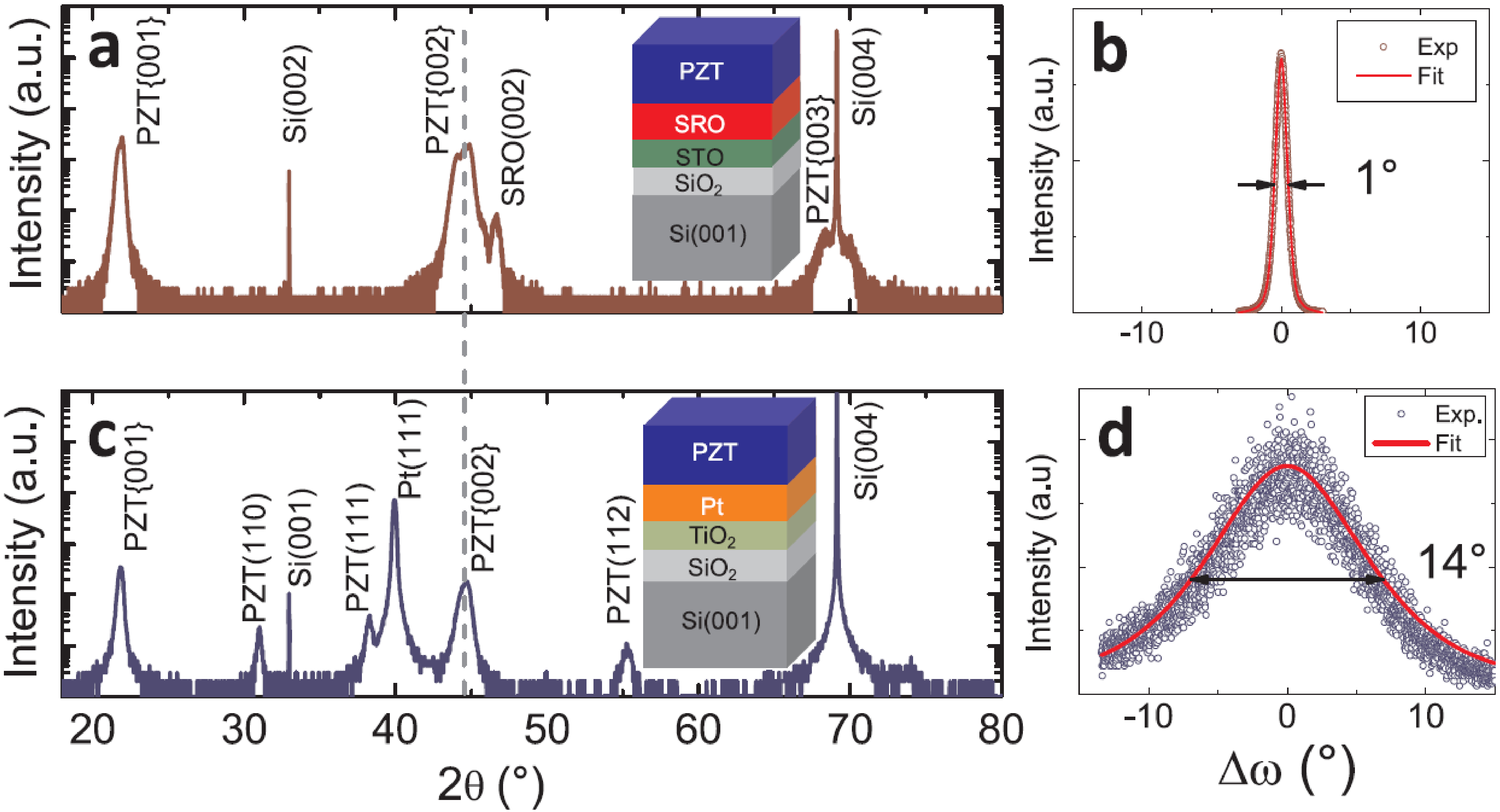
R. Moalla *et al.*, CrystEngComm **18**, 1887 (2016)

Key results from literature

Impact of the epitaxy

Epitaxial

Polycrystalline
(mainly textured)



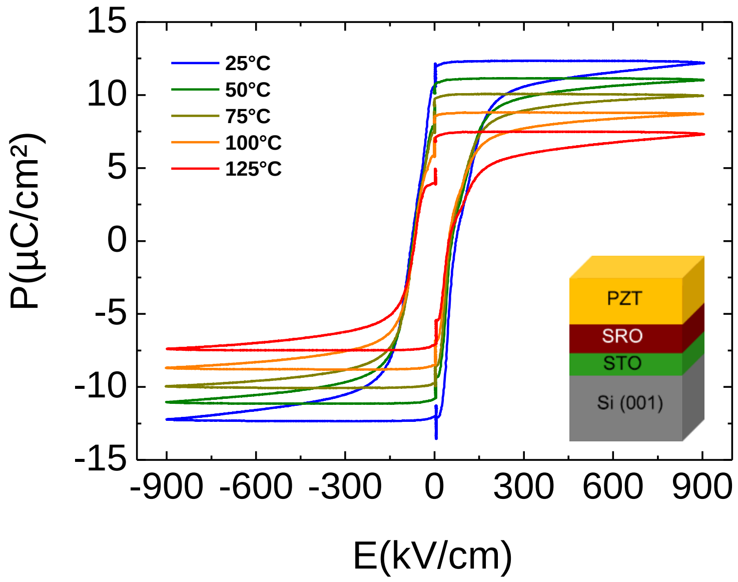
R. Moalla *et al.*, Nano Energy **41**, 43 (2017)

Collab. CEA Leti (E. Defaÿ, G. Le Rhun)

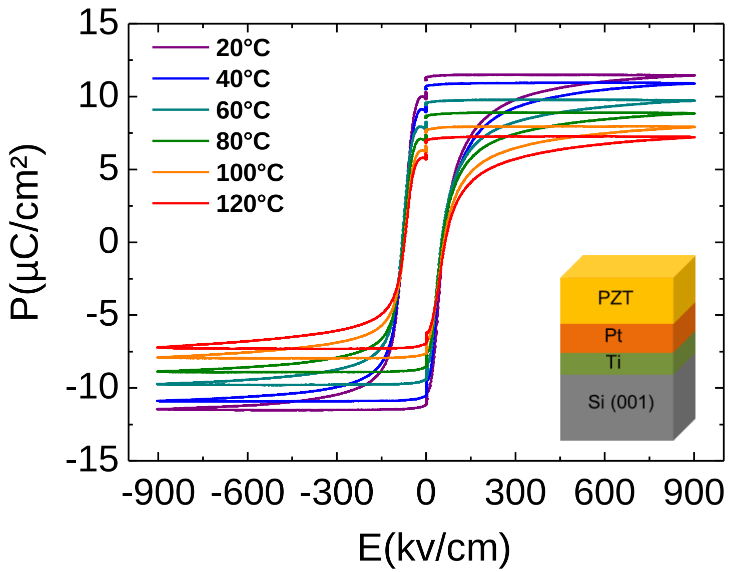
Impact of the epitaxy

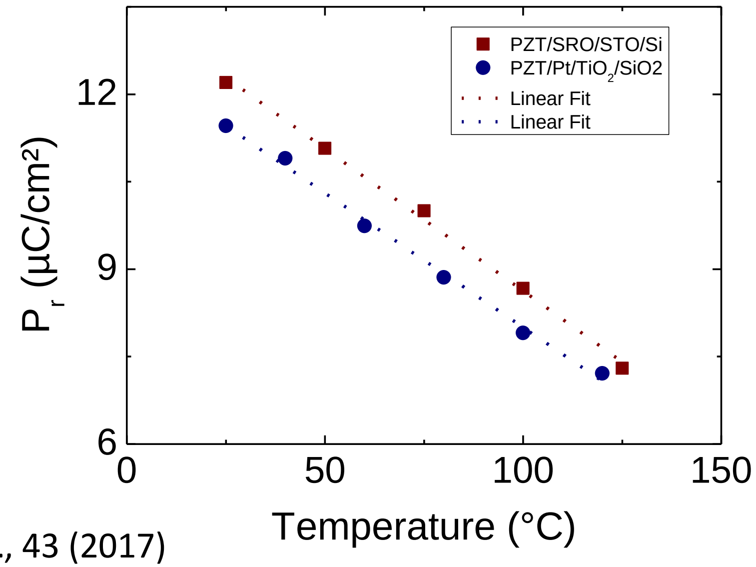
Indirect
(no extrinsic effect)

Epitaxial



Polycrystalline





Pyroelectric coefficients:

$p_{\text{epitaxial}} = -480 \mu C.m^{-2} K^{-1}$

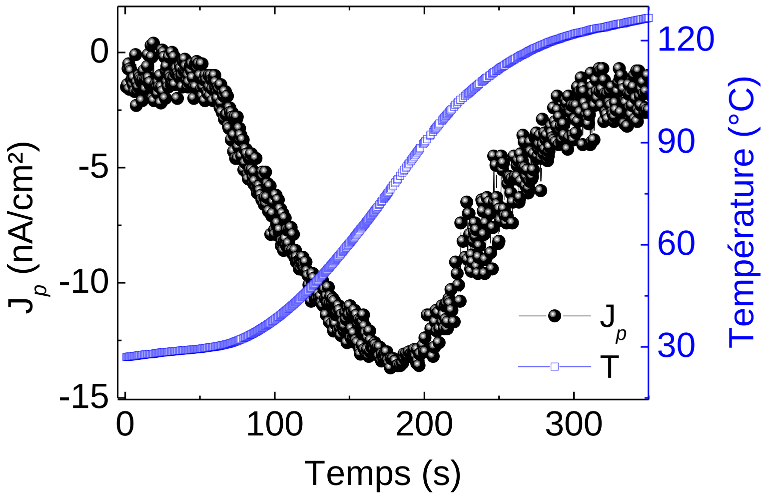
$p_{\text{polycrystalline}} = -460 \mu C.m^{-2} K^{-1}$

R. Moalla *et al.*, Nano Energy **41**, 43 (2017)

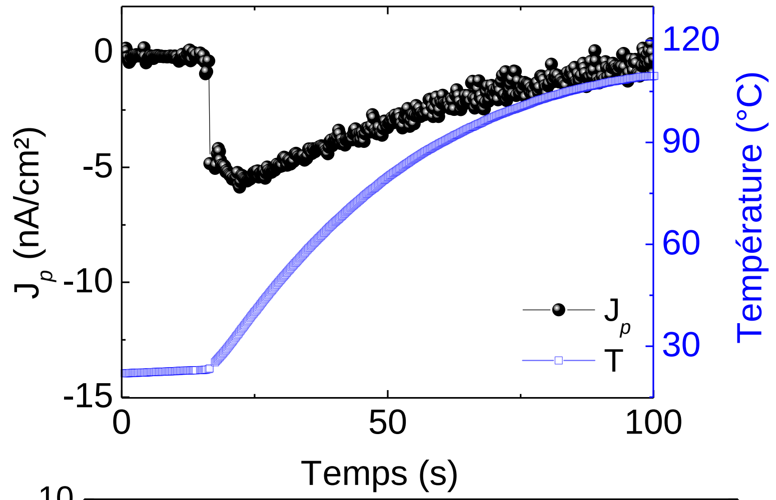
Impact of the epitaxy

Direct
(all contributions)

Epitaxial



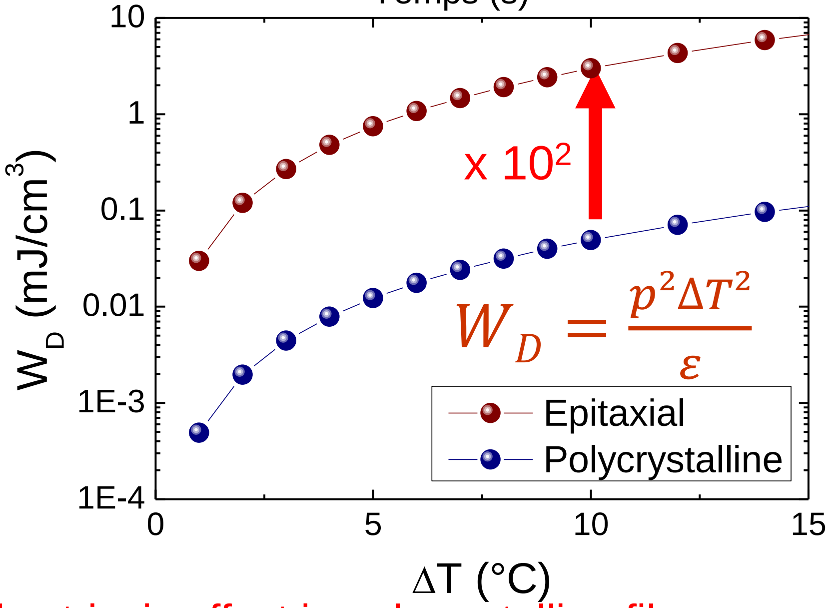
Polycrystalline



$$p = \frac{-i}{A\,\frac{dT}{dt}}$$

$p_{épi} = -220\ \mu C.m^{-2}\ K^{-1}$

$p_{poly} = -26\ \mu C.m^{-2}\ K^{-1}$



2 orders of magnitude!!

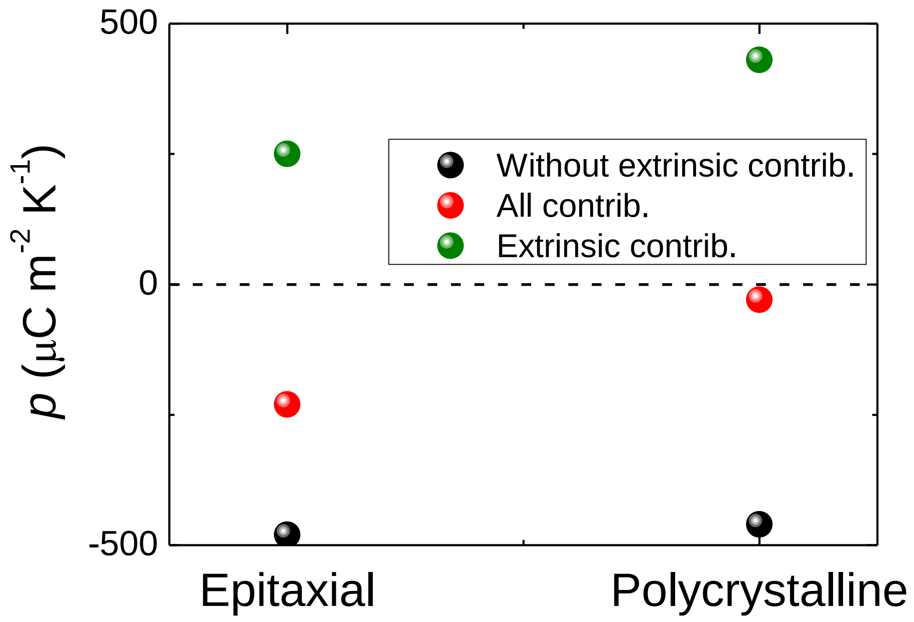
R. Moalla *et al.*, Nano Energy **41**, 43 (2017)

⇒ Large detrimental extrinsic effect in polycrystalline films even textured!

Impact of the epitaxy

Statics (indirect): *without extrinsic effects*
Dynamics (direct): *with extrinsic effects*

⇨ ***Discrimination of the extrinsic effect***



➔ *Measured extrinsic effects: in agreement with theoretical predictions*

J. Karthik et al., PRL **109**, 257602 (2012)

Impact of the substrate (TEC) & strain (c/a preferential orientation)

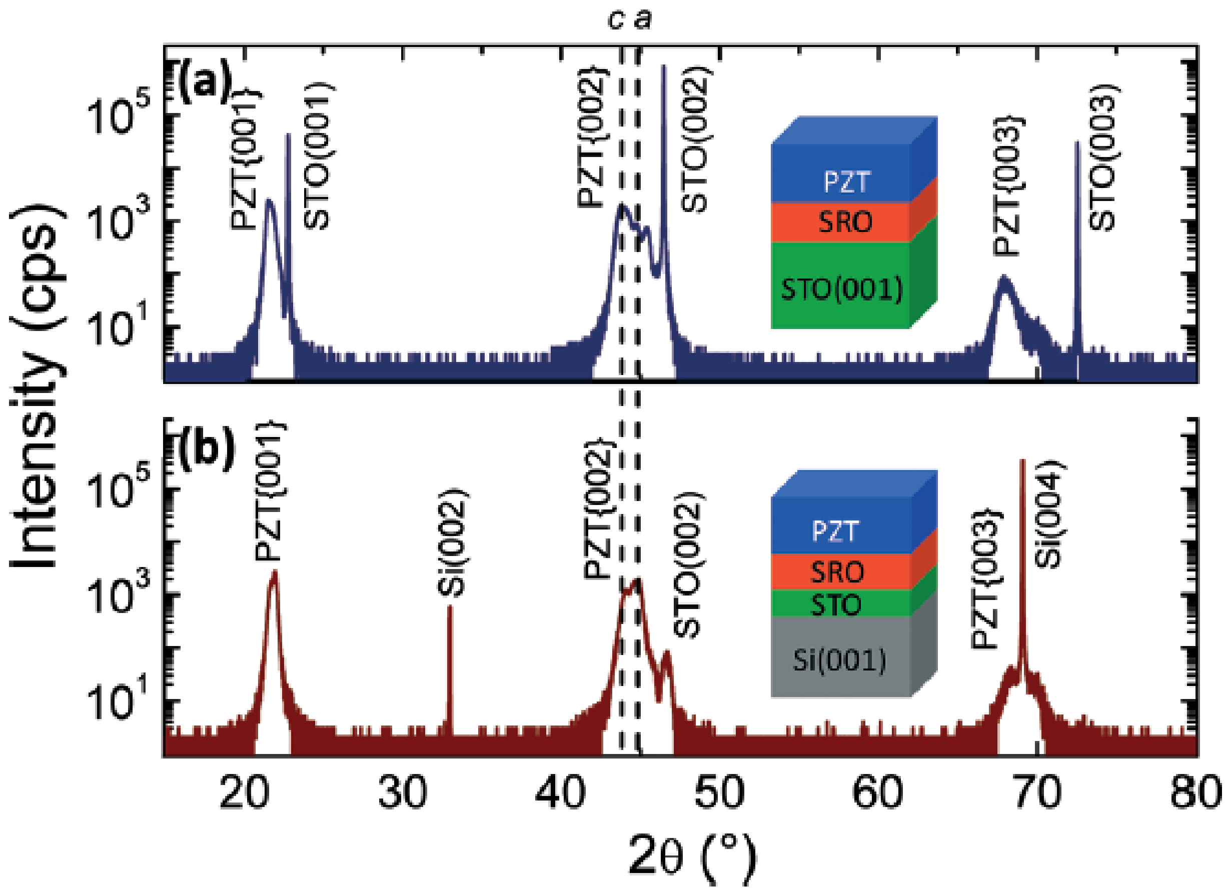
a $_{PZT}$ > a $_{STO}$

TEC $SrTiO_3$ (STO) & PZT = 9 × 10^{-6} K^{-1}

TEC Si = 2.6 × 10^{-6} K^{-1}

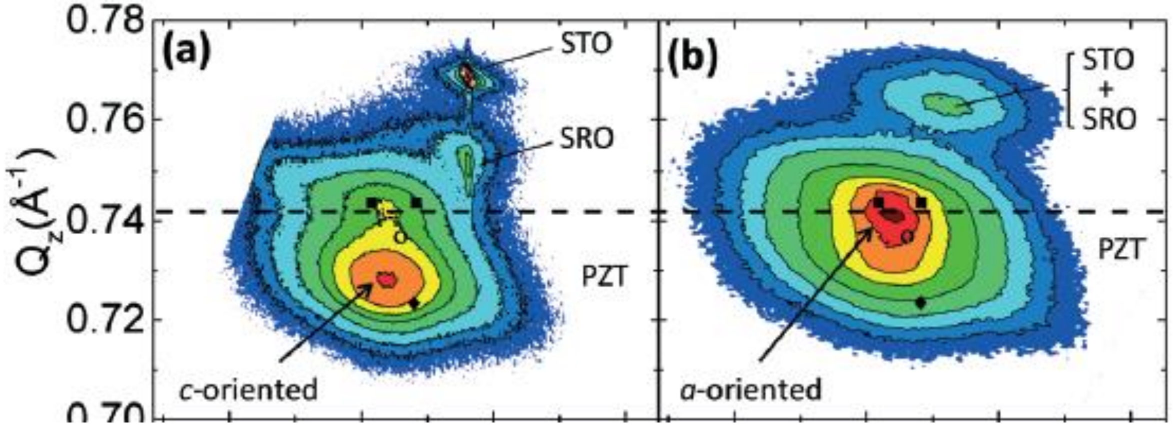
➔ on STO: PZT epitaxially strained in compression in plane

➔ on Si: PZT thermally strained in tension in-plane



/STO

/Si



Mainly ***c***-oriented domains

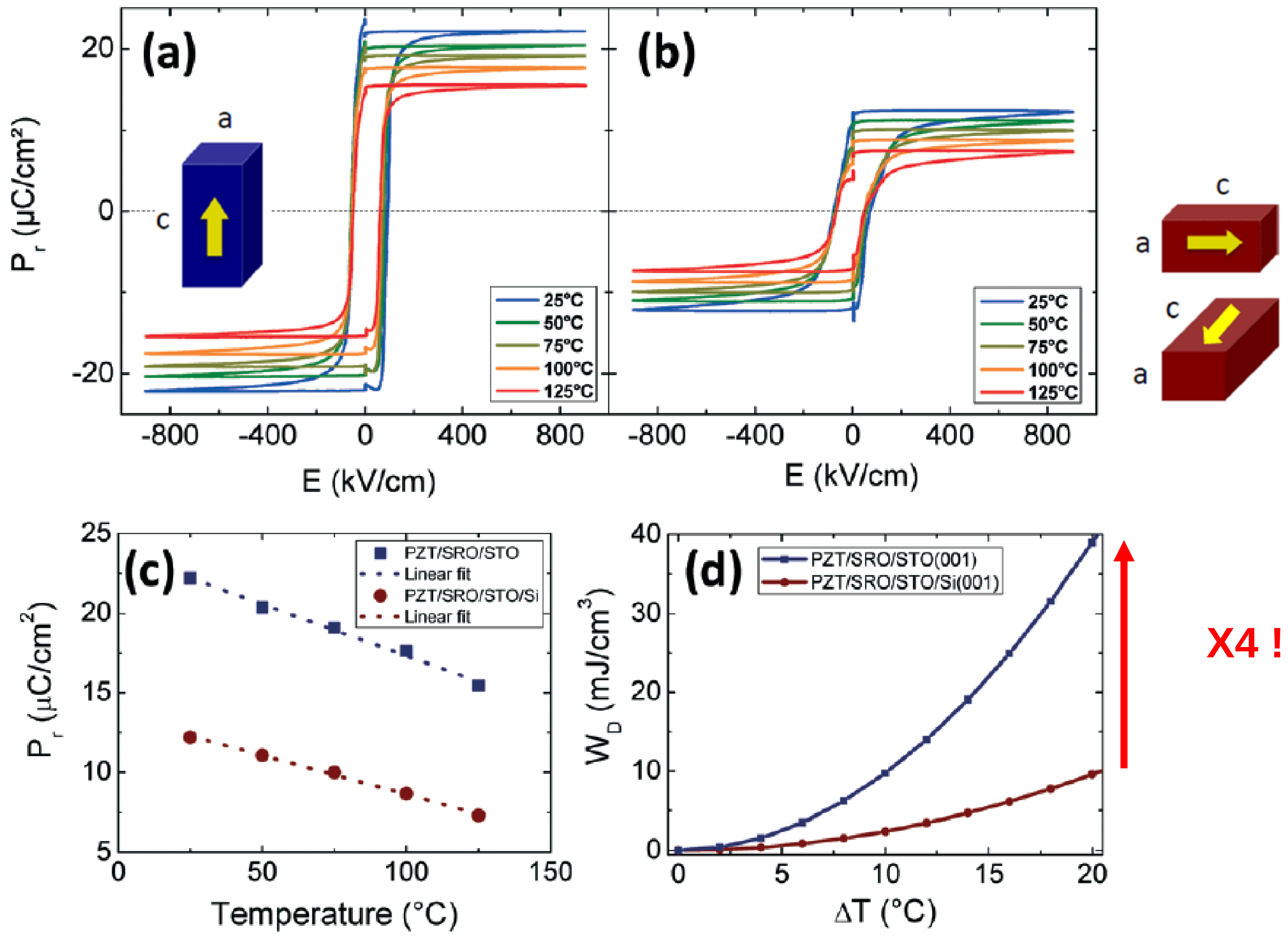
Mainly ***a***-oriented domains

R. Moalla *et al.*, CrystEngComm **18**, 1887 (2016)

Impact of the substrate (TEC) & strain (c/a preferential orientation)

Pr RT /STO = 22.5 µC cm^{-2}
Pr RT /Si = 13.5 µC cm^{-2}

PZT//STO **PZT//Si**

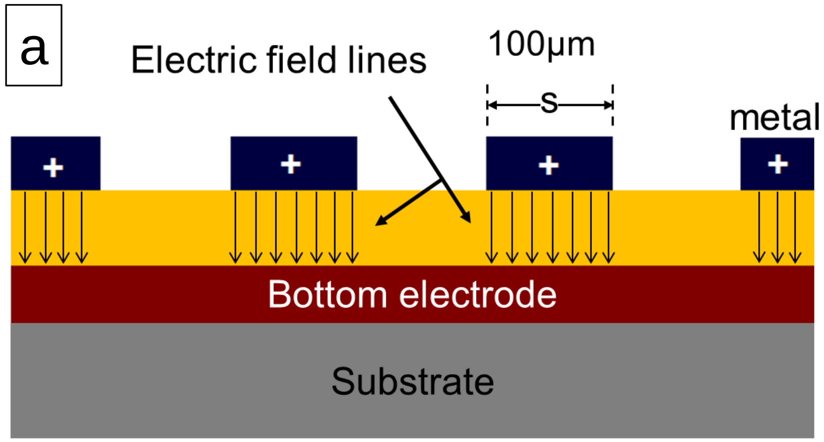


p /STO = −680 µC m^{-2} K^{-1}
p /Si = −450 µC m^{-2} K^{-1}

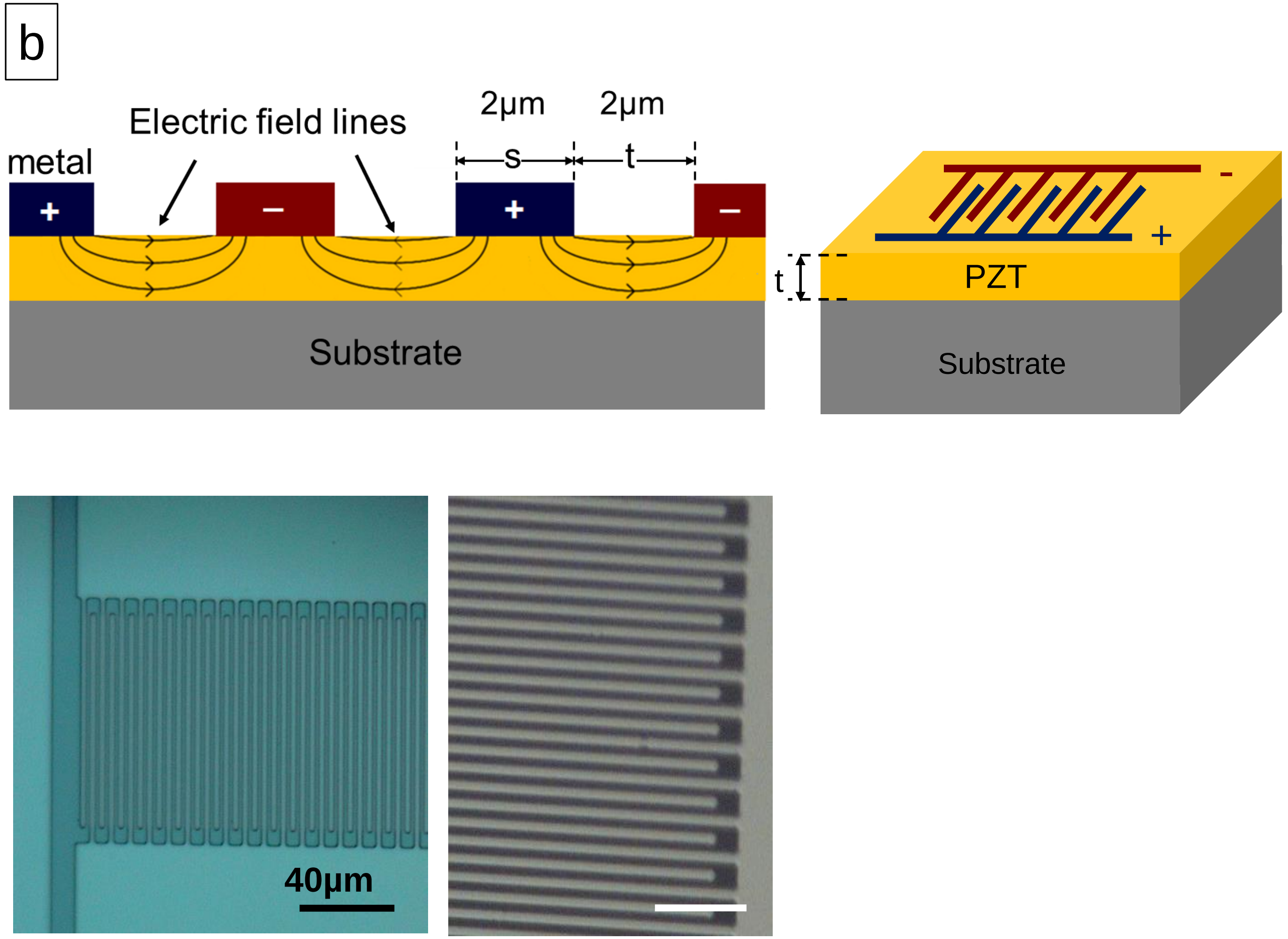
R. Moalla *et al.*, CrystEngComm **18**, 1887 (2016)

Impact of structural anisotropy

Standard out-of-plane measurement

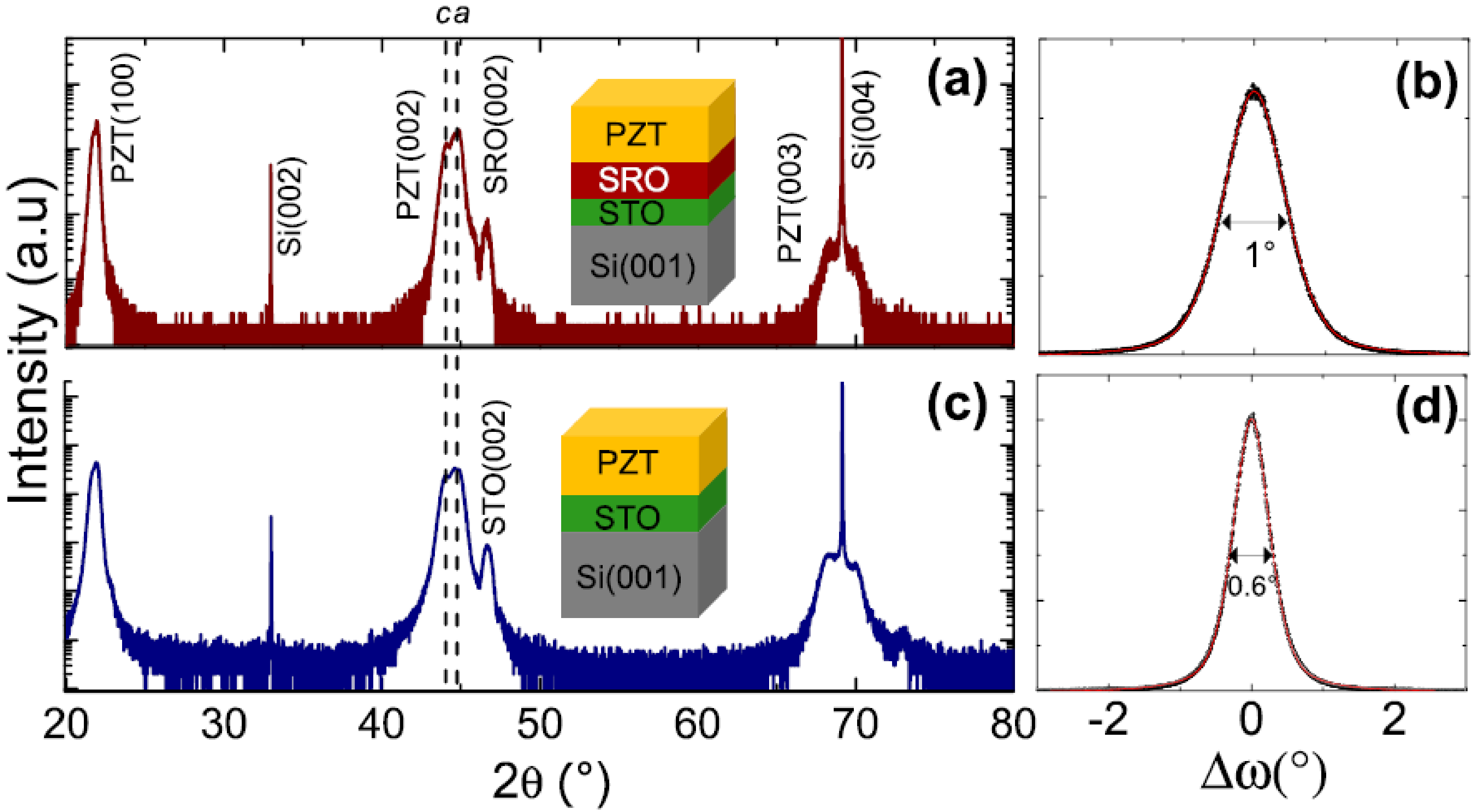


In-plane measurement

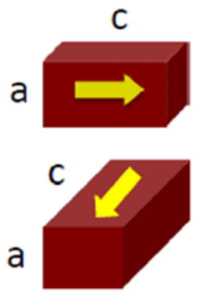


R. Moalla *et al.*, Scientific Reports 8, 4332 **(2018)**

Impact of structural anisotropy

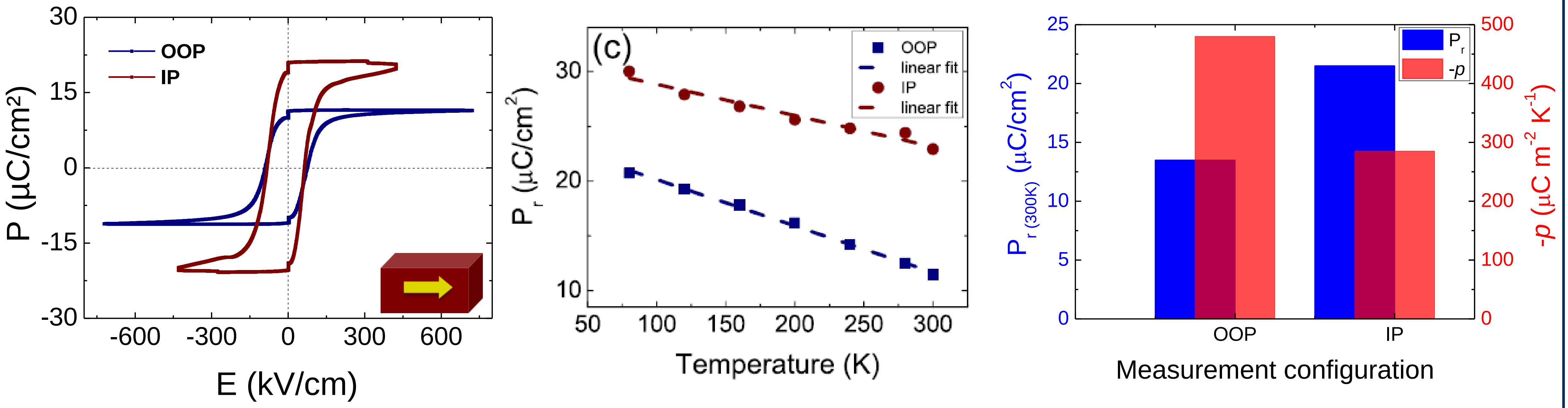


on Si, mainly ***a***-oriented domains for both



R. Moalla *et al.*, Scientific Reports 8, 4332 **(2018)**

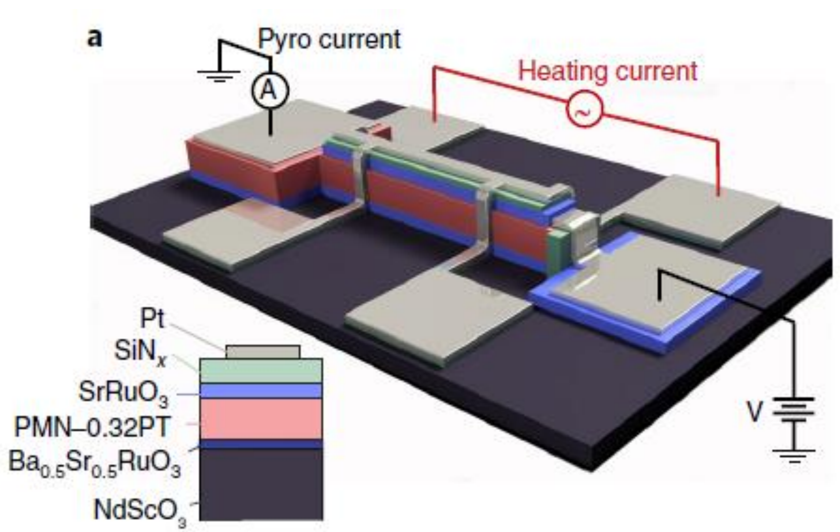
Impact of structural anisotropy

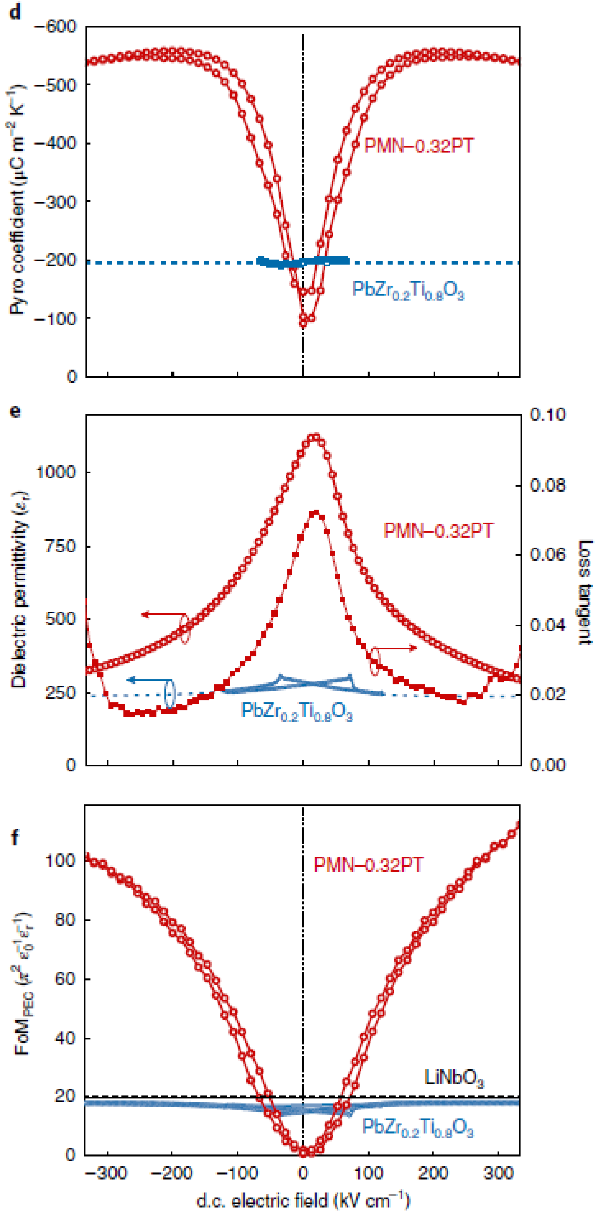


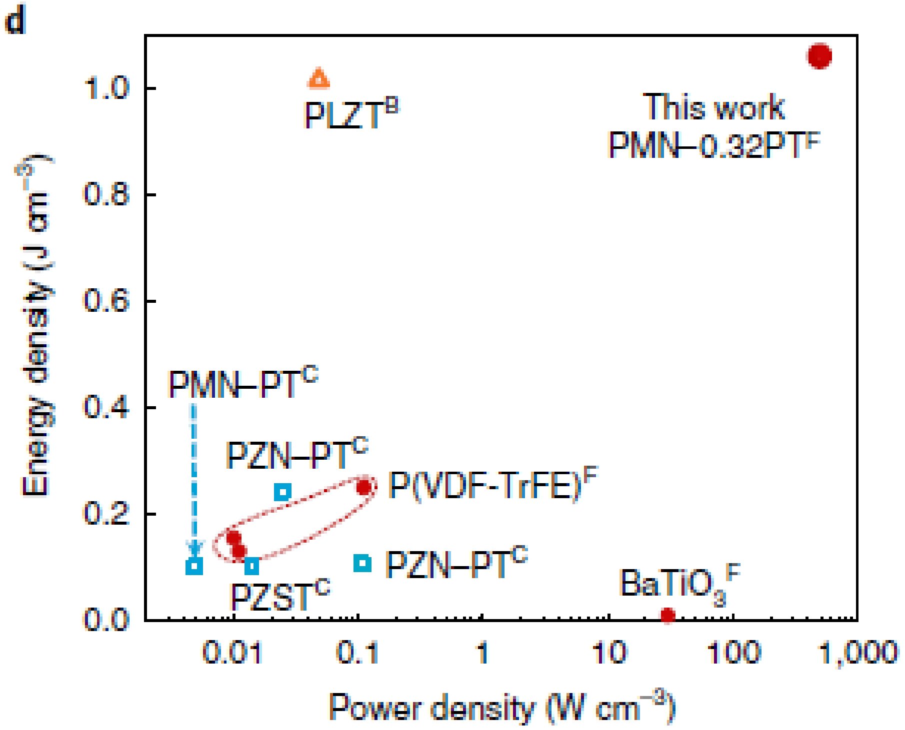
R. Moalla *et al.*, Scientific Reports, 8, 4332 **(2018)**

Enhanced conversions in epitaxial thin films

Using relaxor ferroelectric PMN-PT
And Ericsson cycles







20% Carnot efficiency!

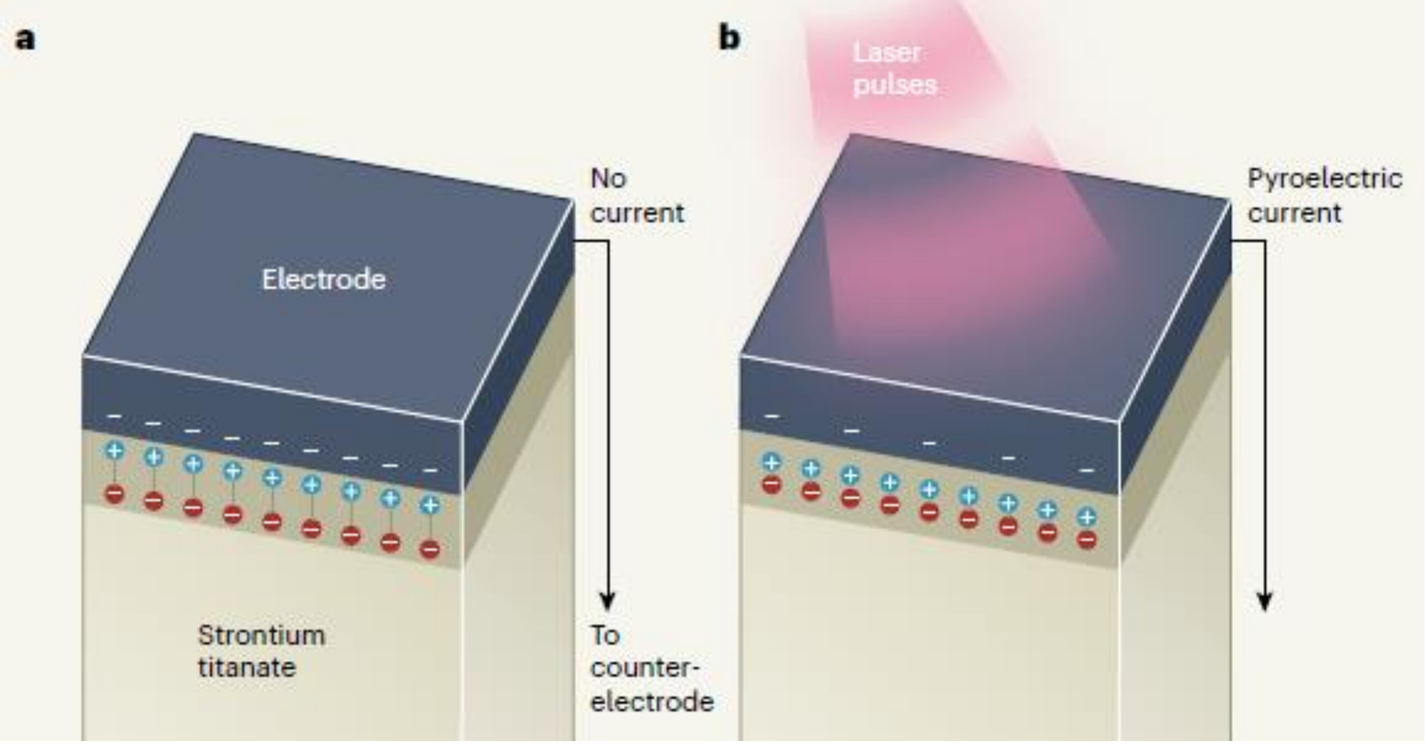
Equivalent to the performance of a thermoelectric with an effective ZT ≈ 1.16 for a temperature change of 10 K !

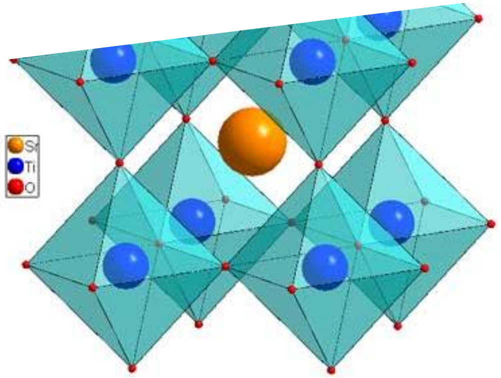
S. Pandya *et al.*, Nature Mater. **(2018)**

Pyroelectricity at surfaces of non-pyroelectric materials

Surface Pyroelectricity in Cubic $SrTiO_3$

By breaking the lattice symmetry





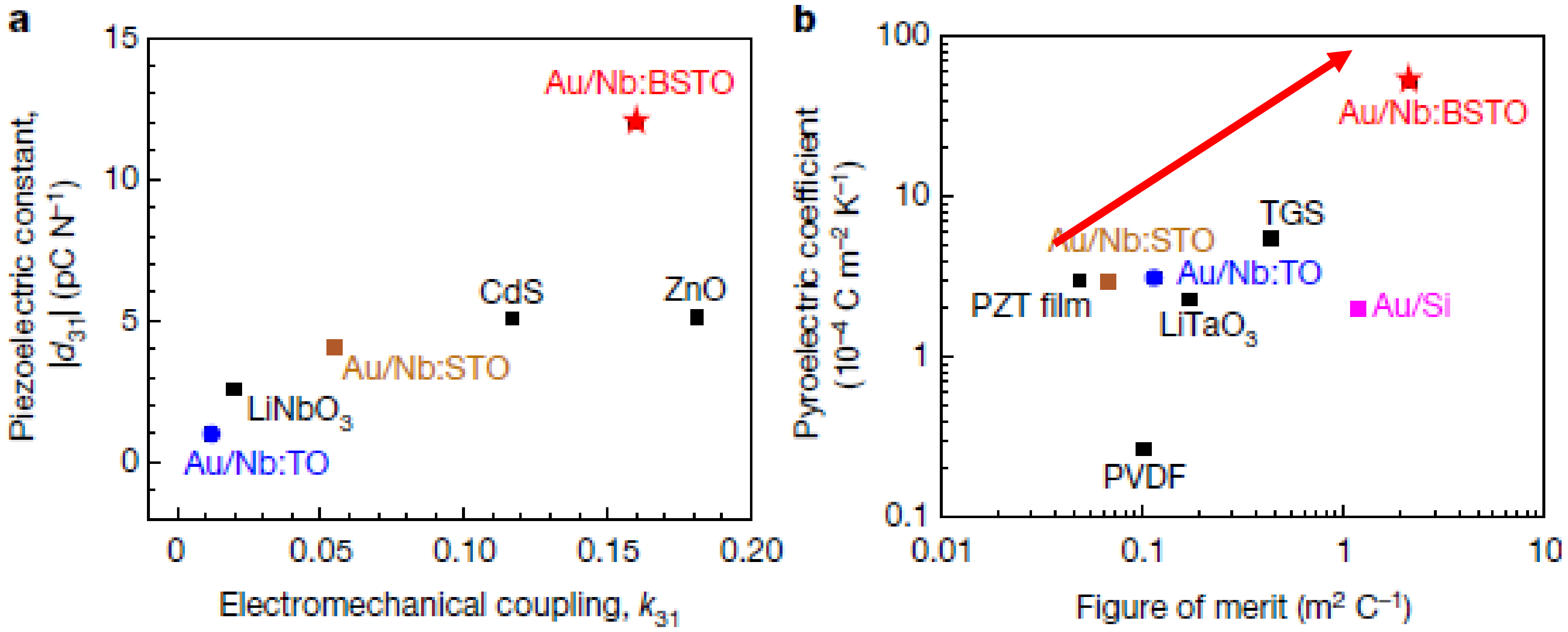
E. Meirzadeh *et al*., Adv. Mater. 1904733 (2019); G. Catalan & B. Noheda, Nature (2020)

Pyroelectricity from interfaces of non-pyroelectric materials

Article

Piezoelectric and pyroelectric effects induced by interface polar symmetry

In centrosymmetric materials!
Band bending at heterointerfaces
➔ Polar symmetry
➔ Built-in E-field



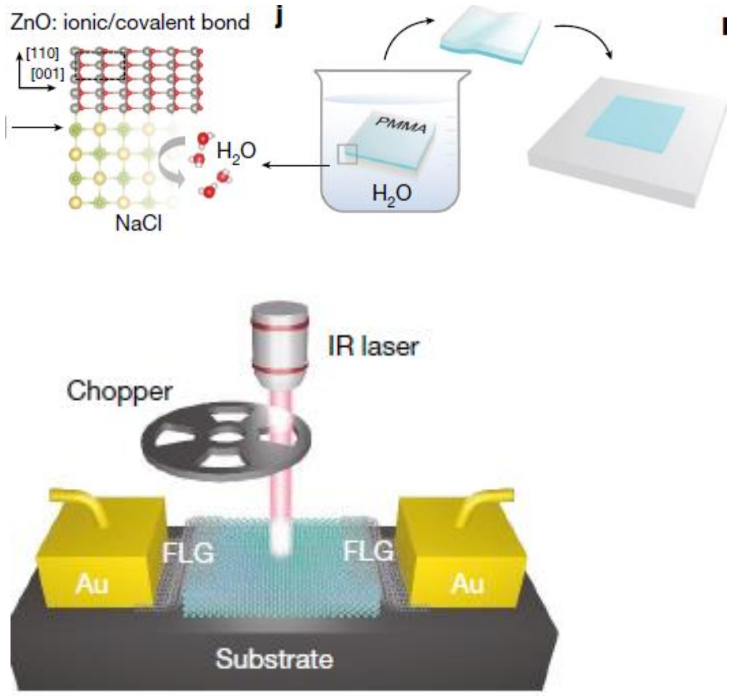
1 order of magnitude than polar pyroelectrics!!

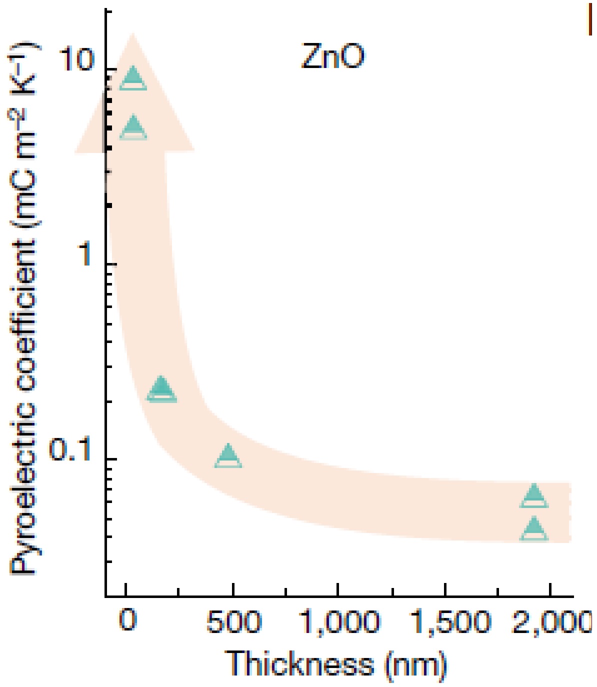
M.-M. Yang *et al.*, Nature 584, 377 (2020)

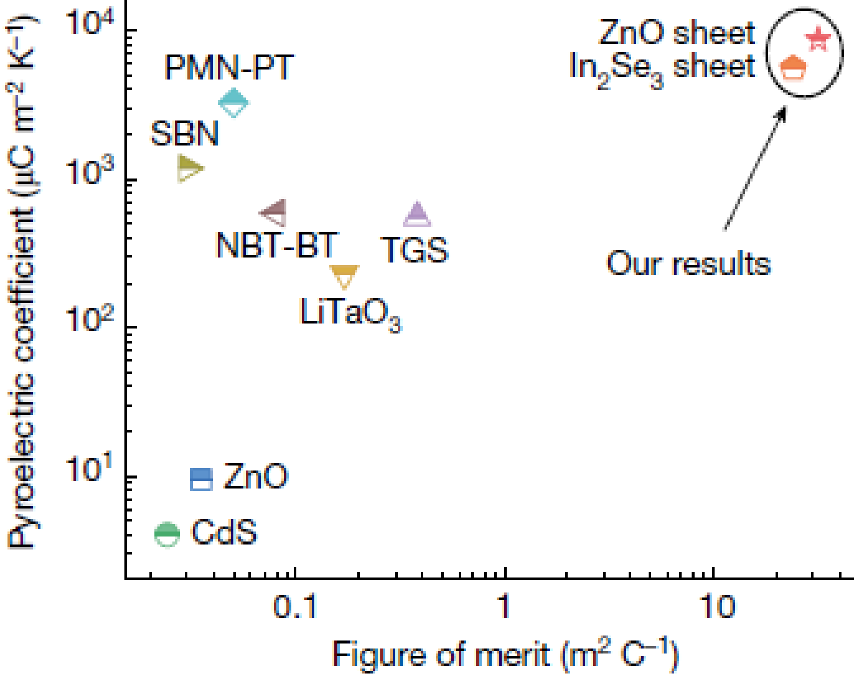
Pyroelectricity in membranes

Article

Giant pyroelectricity in nanomembranes



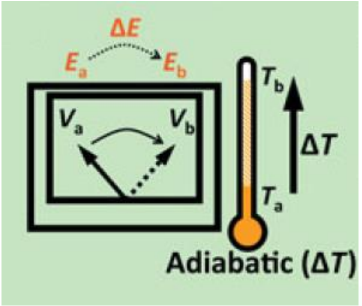


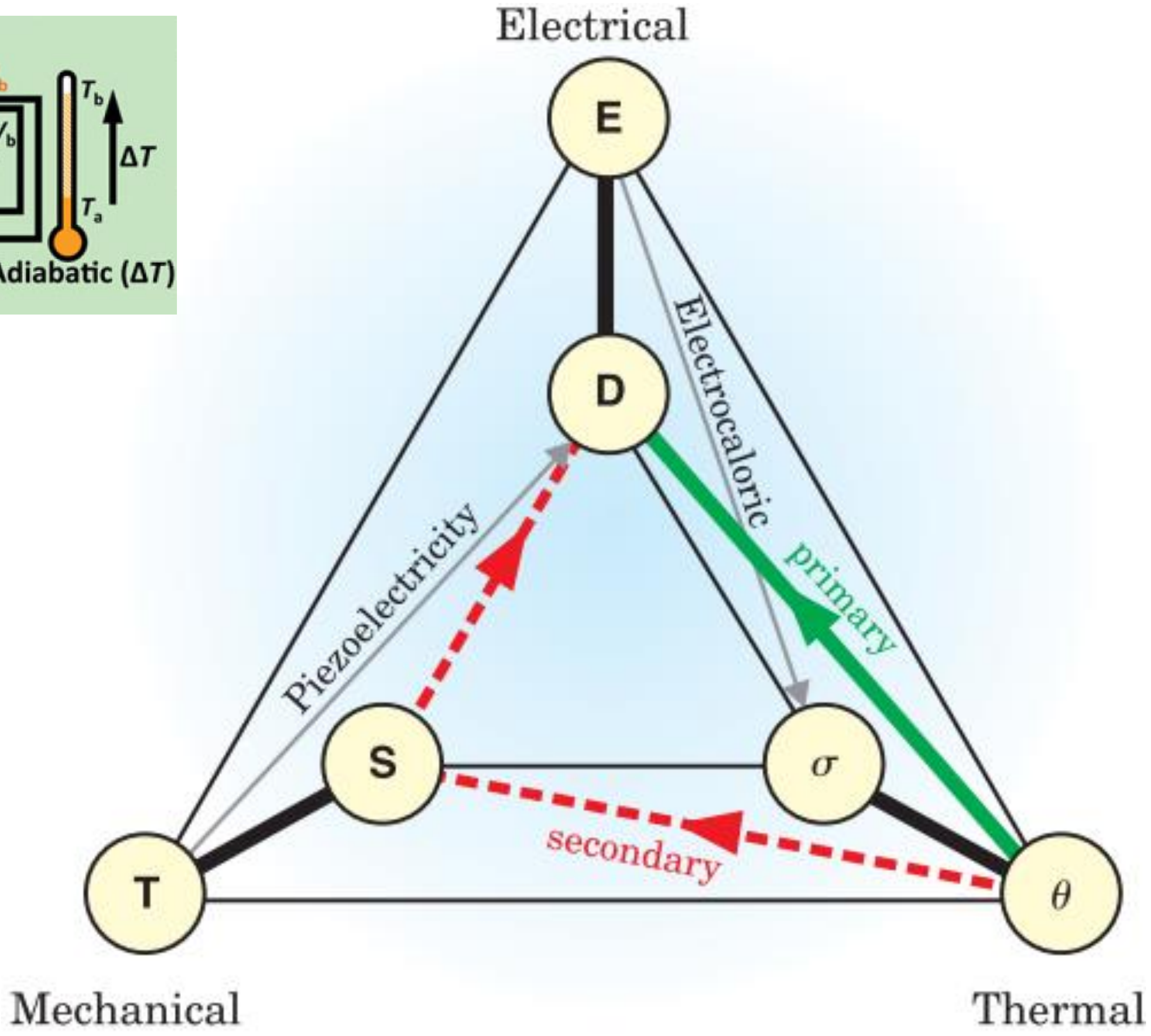


J. Jiang *et al.*, Nature 607, 480 (2022)

Electrocaloric effect (ECE)

Electrocaloric effect: inverse pyroelectric effect in all dielectrics





Heckmann diagram

Polarization variation

Isothermal entropy change (ΔS)

Adiabatic temperature change (ΔT)

Maxwell relation

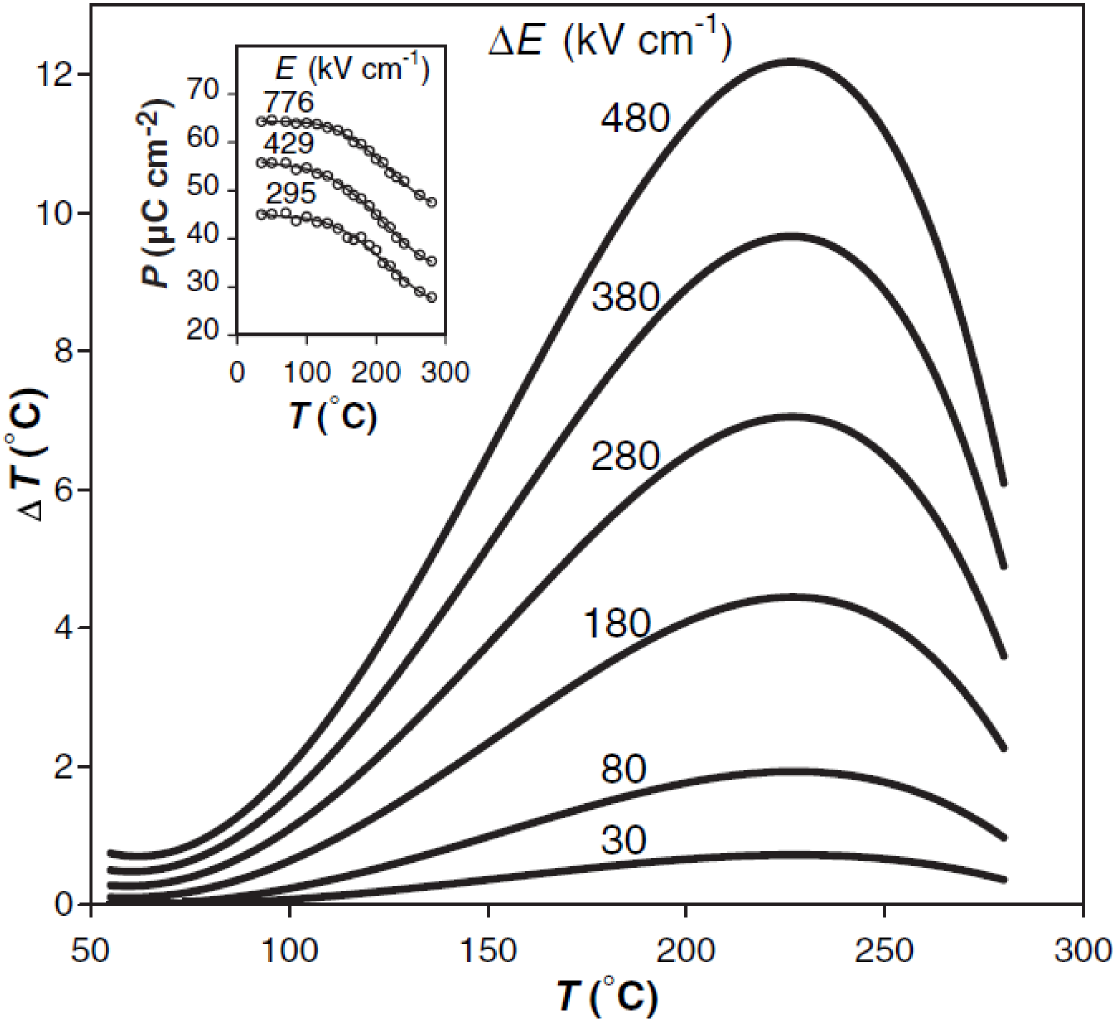
$$\left(\frac{\partial P}{\partial T}\right)_E = \left(\frac{\partial S}{\partial E}\right)_T$$

$$\Delta S = -\frac{1}{\rho}\int_{E_1}^{E_2}\left(\frac{\partial P}{\partial T}\right)_E \mathrm{d}E$$

$$\Delta T = -\frac{1}{C\rho}\int_{E_1}^{E_2} T\left(\frac{\partial P}{\partial T}\right)_E dE$$

Electrocaloric effect: inverse pyroelectric effect in all dielectrics

Giant electrocaloric effect (ECE) in PZT (95/05) – measured indirectly from P-E & equations



Other caloric materials:
See the reviews
X. Moya *et al*., Nat. Mat. **13**, 439 (2014)

Also
X. Moya *et al*., Nat Phys. **11**, 202 (2015)

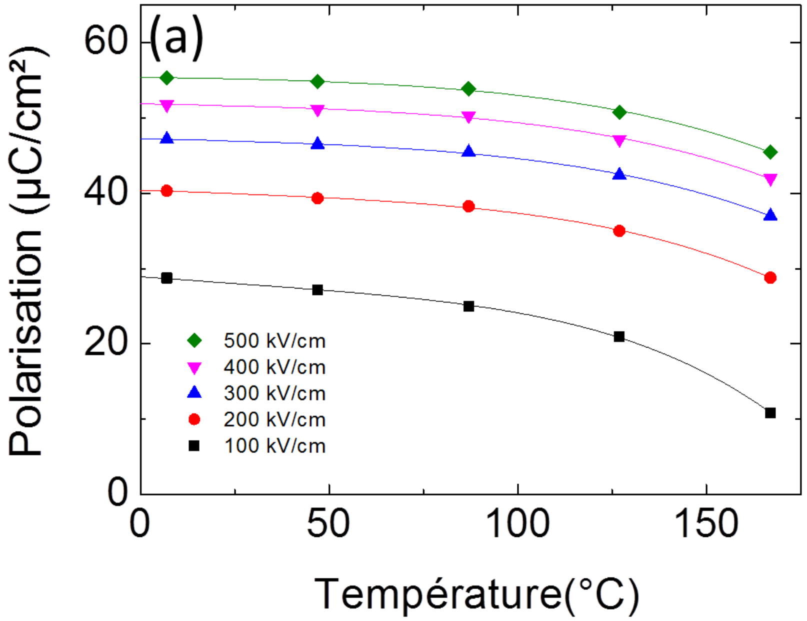
A.S. Mischenko *et al.*, Science **311**, 1270 (2006); G. Sebald et al., JAP **100**, 124112 (2006)

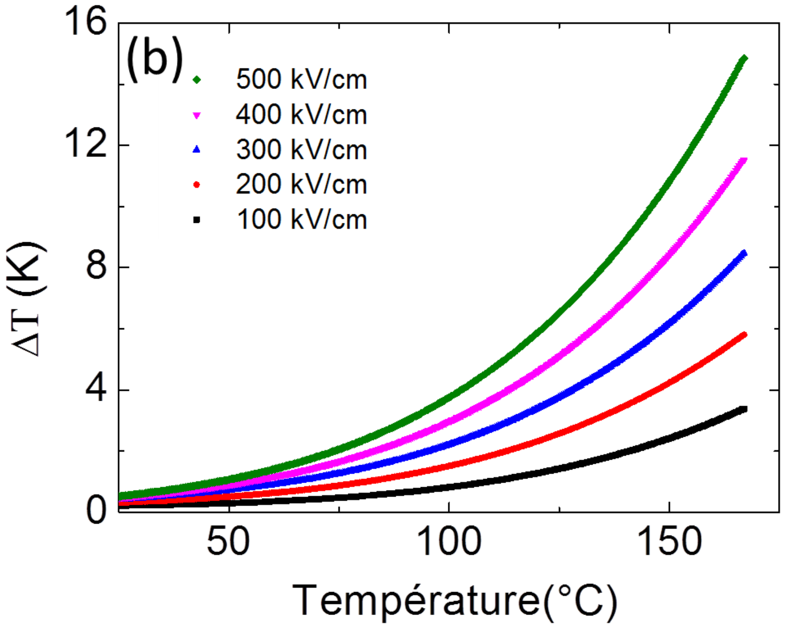
Electrocaloric effect: inverse pyroelectric effect in all dielectrics

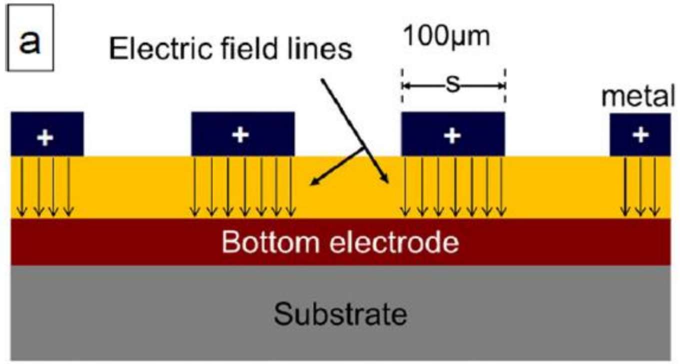
Electrocaloric effect for cooling applications

Same indirect measurements done @INL

***0.5 μm** PZT (52/48) on STO(001)*







$$\Delta T = 15\ K\ @\ E = 500\ kV/cm$$

R. Moalla *et al.*, PhD Thesis INL-ECL (2016)

Electrocaloric effect: inverse pyroelectric effect in all dielectrics

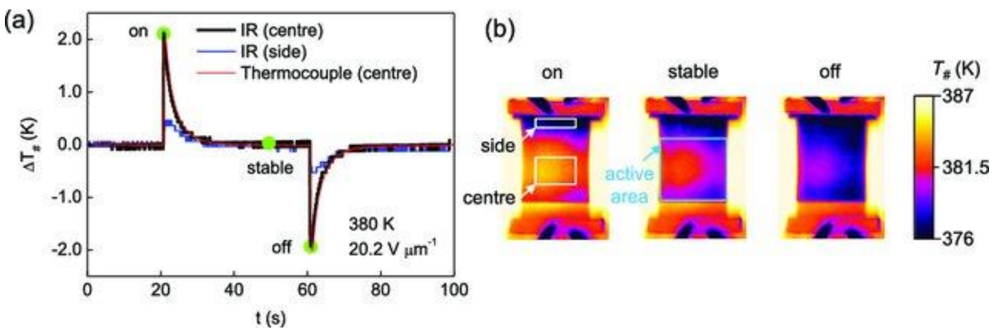
Electrocaloric effect for cooling applications

Review: Y. Liu *et al*., Appl. Phys. Rev. 3, 031102 (2016)

Direct measurements complicated…

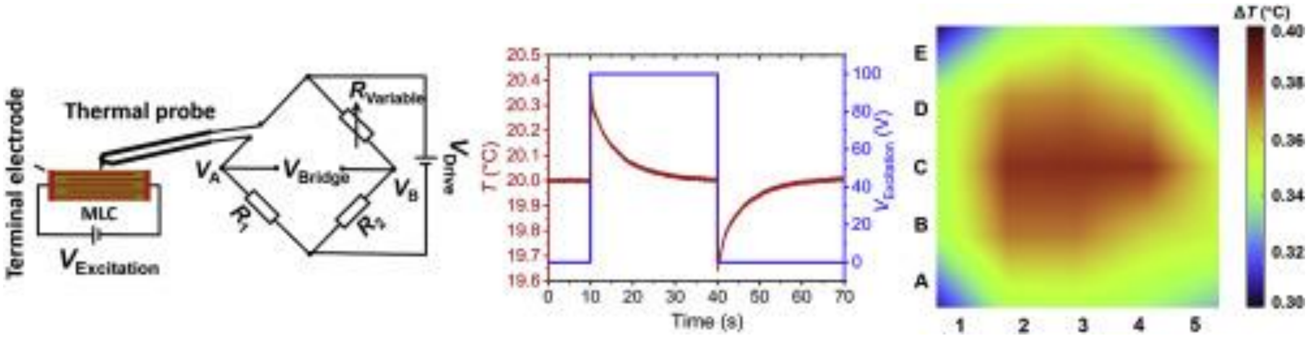
Done mainly on thick multi-layered capacitors (MLCs)

IR camera (but "slow")



T. Usui et al., J. Phys. D (2017)

SThM



D. Shan et al., Nano Energy (2020)

Conclusions & perspectives

Main conclusions

- **Pyroelectrics** as a promising alternative for **thermal energy harvesting**
- Possibility of **cooling applications** from **electrocaloric effect**
- Impact of epitaxy, substrate (strain), domain orientation, anisotropy, E fields,…

Perspectives/challenges

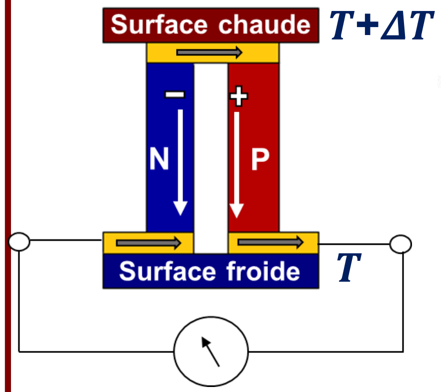
- **Enhanced materials** (Pb-free, epitaxy on Si, phase change boundaries,…)
- **On-chip microsystems** (oscillators, hybrids,…)

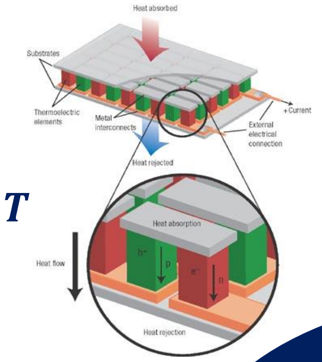
Thermoelectricity (TE) *vs* Pyroelectricity (PE)

TE

Thermoelectrics

ΔT → i





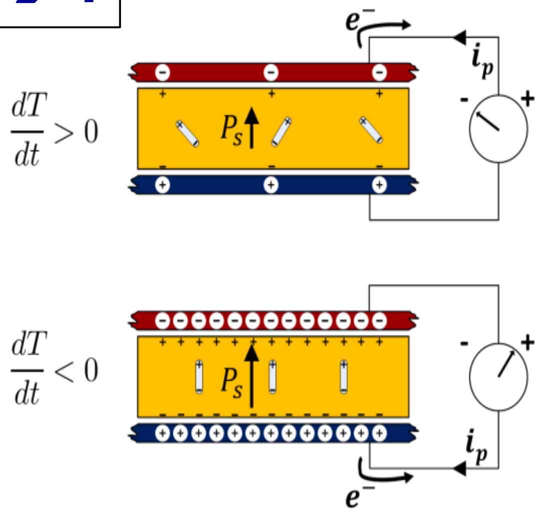
- Thermal difference
- 2 materials (*n* et *p*)
- Nanostructuration

Thermal energy harvesting

PE

Pyroelectrics

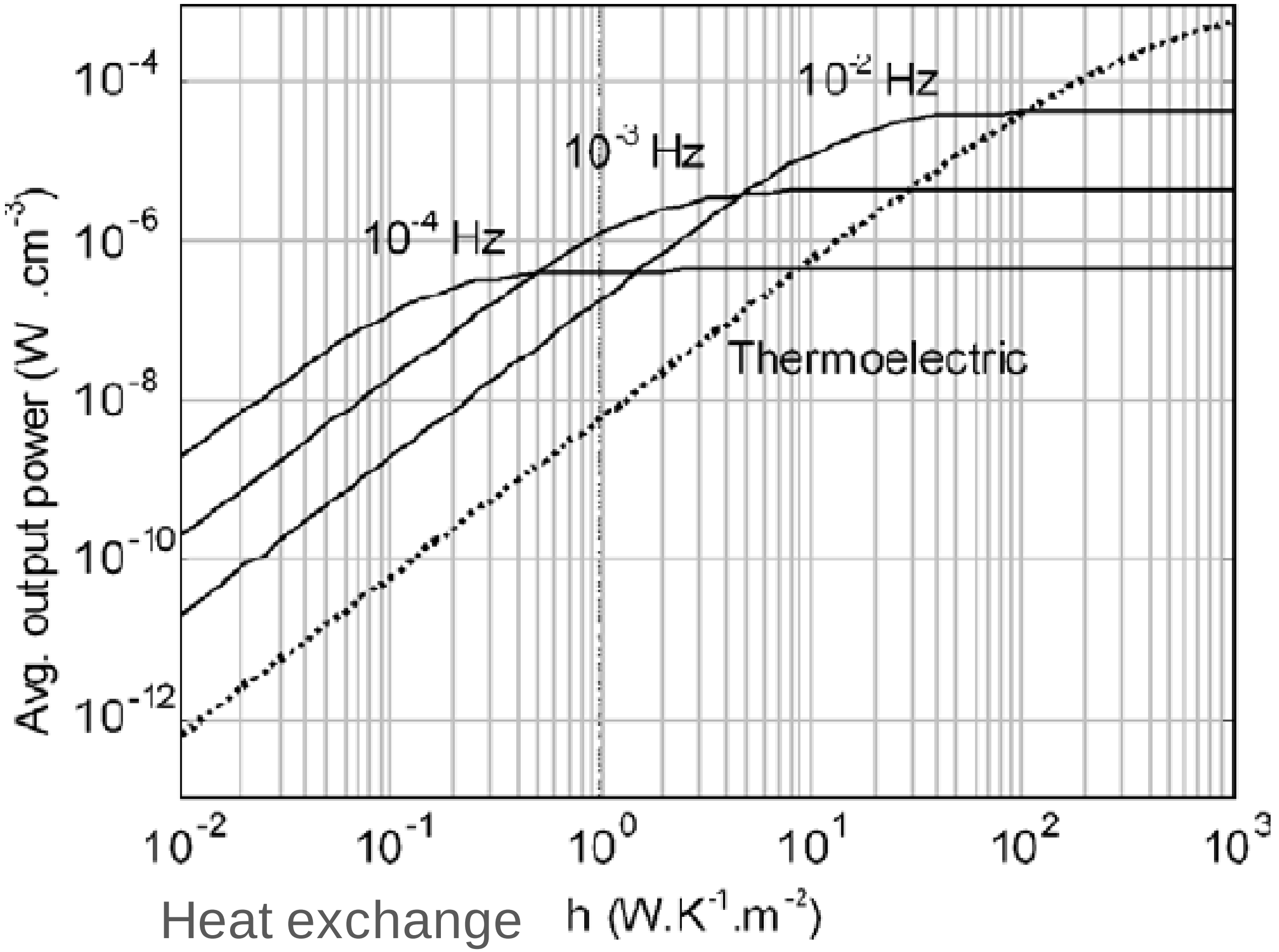
dT/dt → i



- Temporal change of T
- Can be thin films
- No structuration

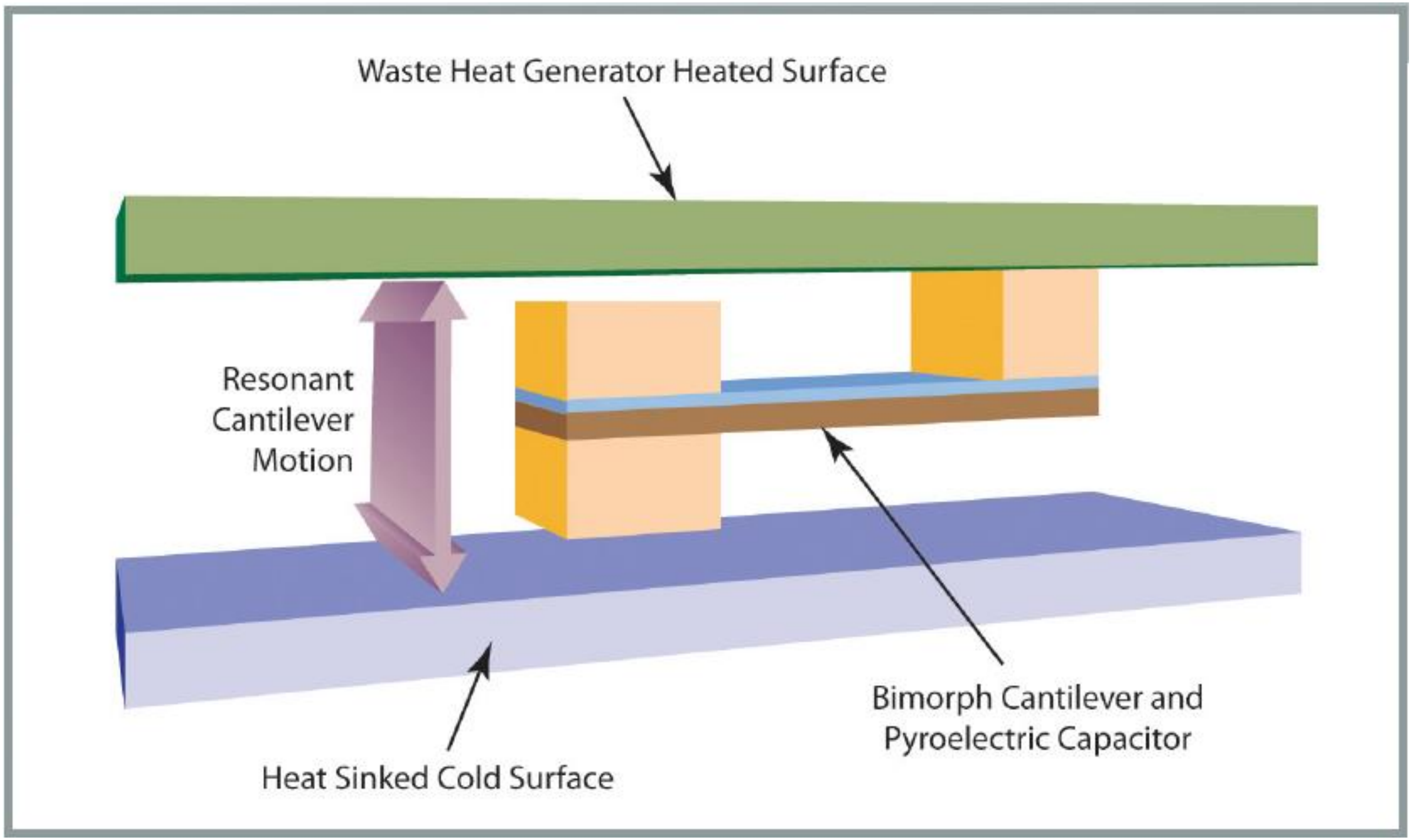
Thermoelectricity (TE) *vs* Pyroelectricity (PE)

Comparable output power



G. Sebald *et al.*, Smart Mater. Struct. **18**, 125006 (2009)

Artificially higher rate of T variations



C. Bowen *et al.*, En. Env. Sci. **(2014);** *S.R. Hunter, USA Patent*

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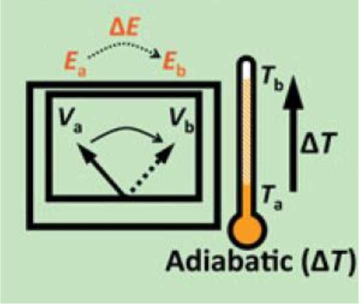

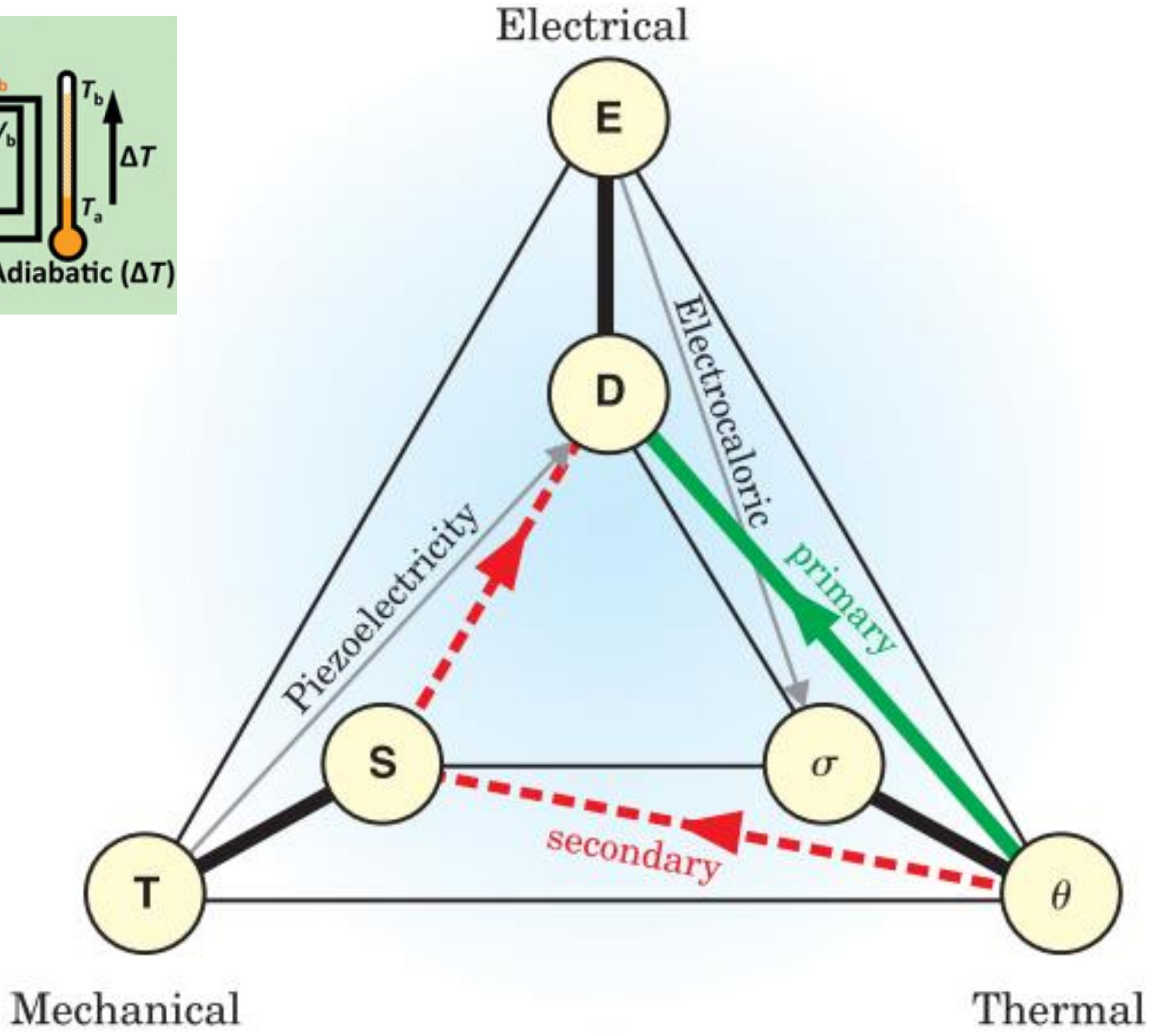

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⬇

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➕

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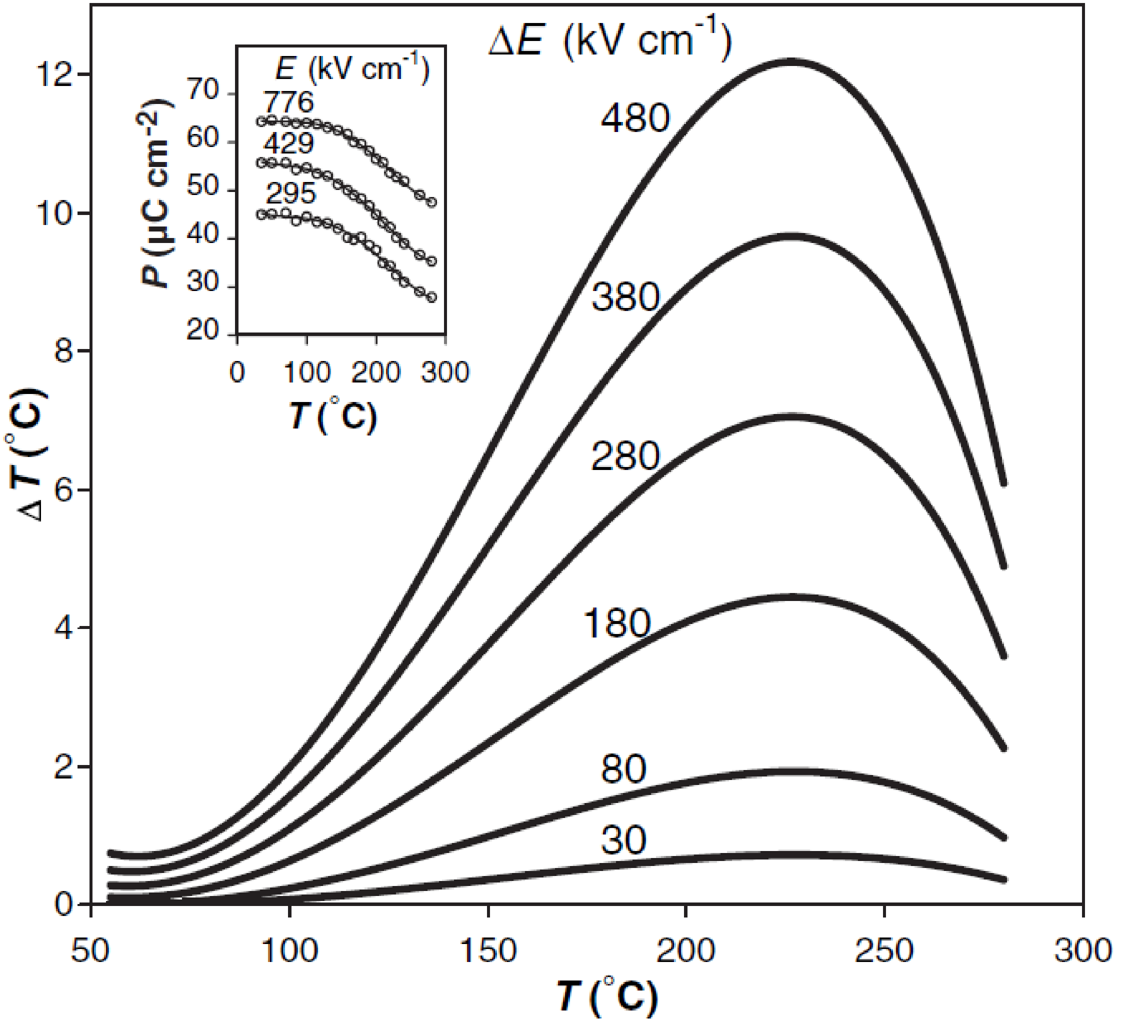

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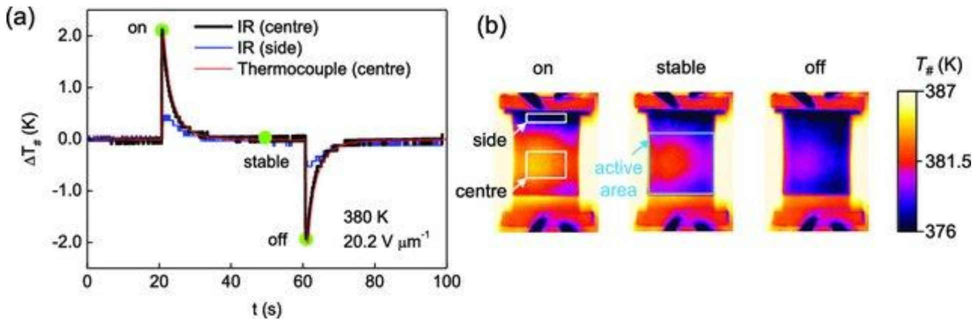
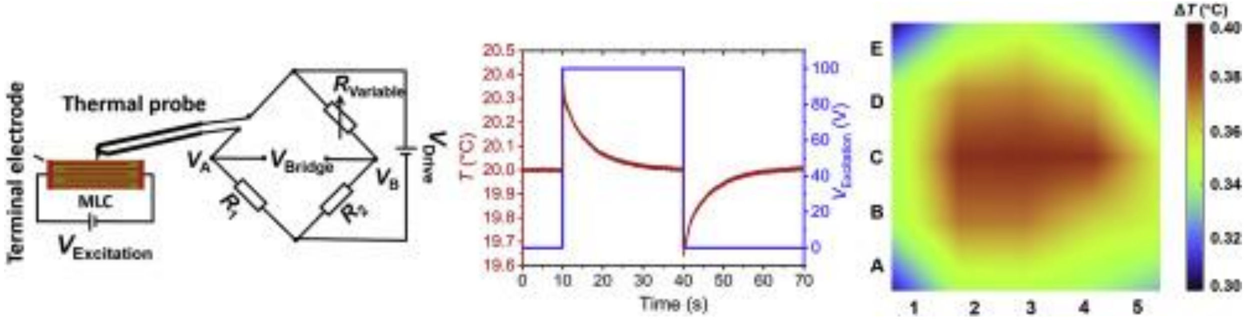


T. Usui et al., J. Phys. D (2017)

SThM



D. Shan et al., Nano Energy (2020)

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C.R. Bowen *et al*., Energy Env. Sci. 7, 3836 (2014)
S.P. Alpay *et al.*, MRS Bulletin 39, 1099 (2014)
G. Velarde *et al.*, APL Mater. 9, 010702 (2021)

Key articles
Y. Yang *et al*., Nano Letters 12, 6408 (2012)
Karthik *et al*., PRL 109, 257602 (2012)
R. Moalla *et al*., Nano Energy 41, 43 (2017)
S. Pandya *et al*., Nature Mater. 17, 432 (2018)
E. Meirzadeh *et al*., Adv. Mater. 1904733 (2019)
M.-M. Yang *et al*., Nature 584, 377 (2020)
N. Mathur’ group (caloric materials)

Des oxydes fonctionnels pour la récupération d’énergie
R. Bachelet, Ouvrage « *L’ingénierie se met au vert* », CNRS Editions (2019)

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Main collaborators on pyroelectrics

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(Oxide MBE, INL)

[redacted]

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(Elec charac., INL)

Gaël Sebald
(LGEF, ELyTMaX)

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CNRS-INSIS PEPS-Energie
ECL BQRs