From Green Piezoelectric Materials to Designed Hybrid Piezoelectric Energy Harvesters

Ausrine Bartasyte*, Micky Rakotondrabe, Giacomo Clementi, Stefania Oliveri, Vincent Astié, Sabina Kuprenaite, Samuel Margueron, and Bernard Dulmet

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MSCN-ETN-722496-ENHANCE

Piezoelectric Energy Harvesters for Self-Powered Automotive Sensors: from Advanced Lead-Free Materials to Smart Systems

Beneficiaries

UFC Université de Franche-Comté

Imperial College London

INSA Institut National des Sciences Appliquées Lyon

Universität zu Köln

INSTM

Grenoble INP

AIXTRON

CEDRAT TECHNOLOGIES Innovation in Mechatronics

13 PhD students

Partner organisations

PSA Groupe

EpiValence Building Chemical Bonds

EPFL École Polytechnique Fédérale de Lausanne

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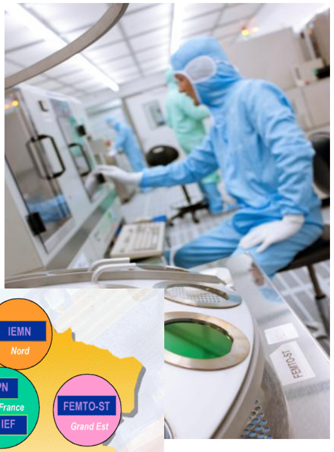
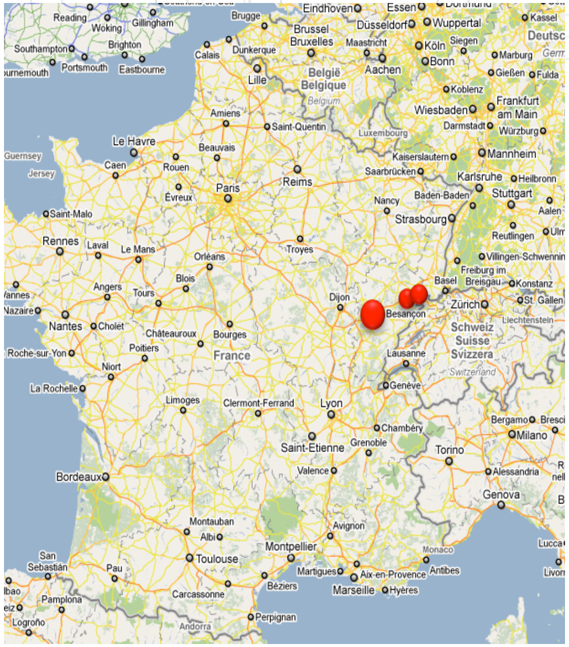
ST

FEMTO-ST Institute: long standing expertise in electroactive applications & $LiNbO_3$

- **The Biggest Institute in Engineering in France** (700 members including 325 permanent staff, 125 non-permanent staff and 250 PhD students).
- **Clean Room MIMENTO**: one of the biggest clean room facilities in France

(800 m², equipments for 13 M€,
pilot line for industrial microfabrication
of acoustic devices)





7 research departments:

TIME-FREQUENCY

OPTICS

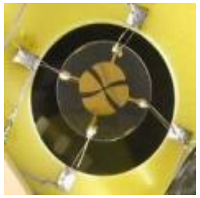
MICRO NANO SCIENCES & SYSTEMS (MN2S)

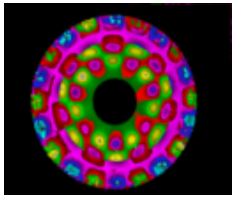
APPLIED MECANICS

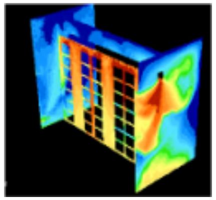
AUTOMATICS & MICRO-MECATRONIC SYSTEMS

ENERGY

INFORMATICS OF COMPLEX SYSTEMS







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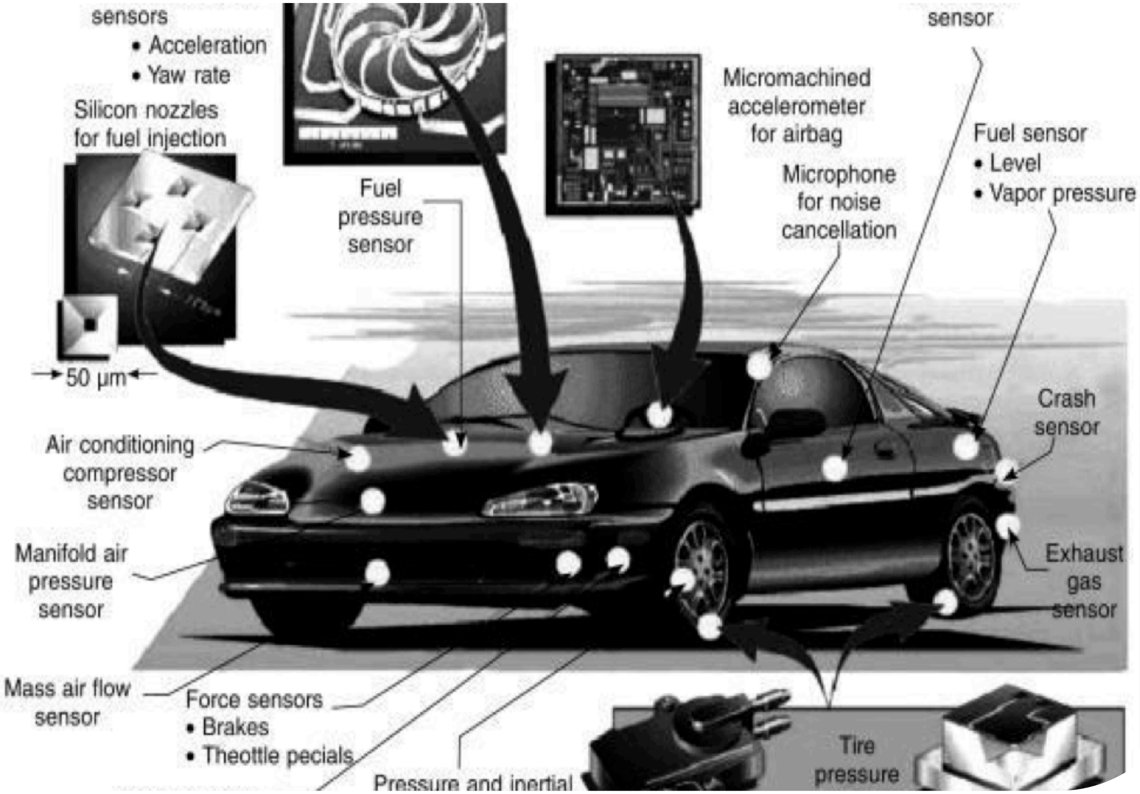
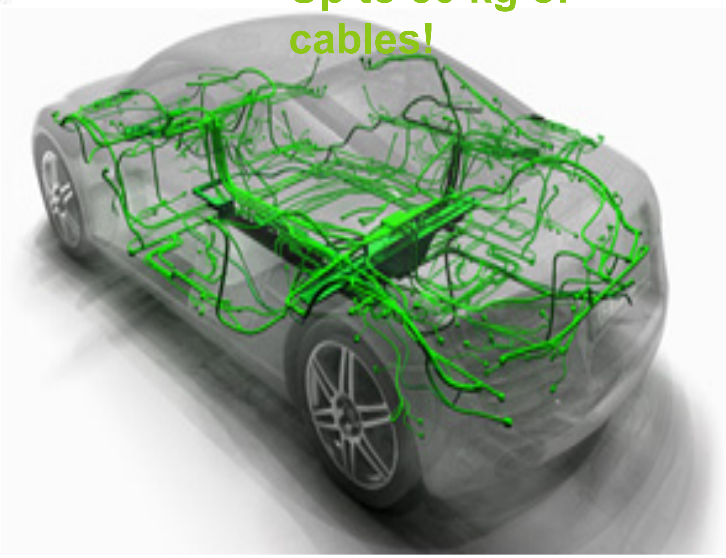
Automotive/aerospace sensors



Automotive MEMS (Precision Engineering By V. C. Venkatesh, Sudin Izman)

Challenges: weight, complexity, cost of maintenance, sensibility to corrosive environment

Up to 50 kg of cables!



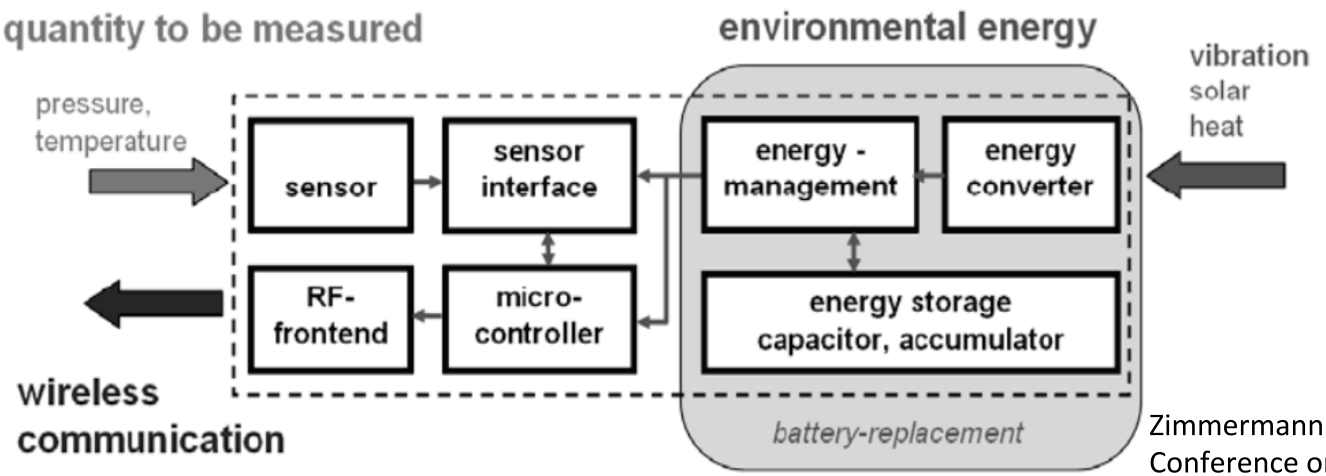
Future vehicles:

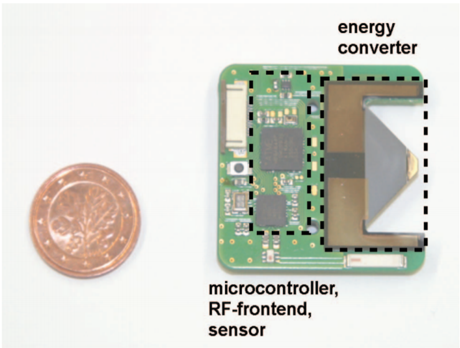
- Autonomous, wireless & maintenance-free MEMS sensors

Automotive/aerospace sensors

Future vehicles:

- Autonomous, wireless & maintenance-free MEMS sensors

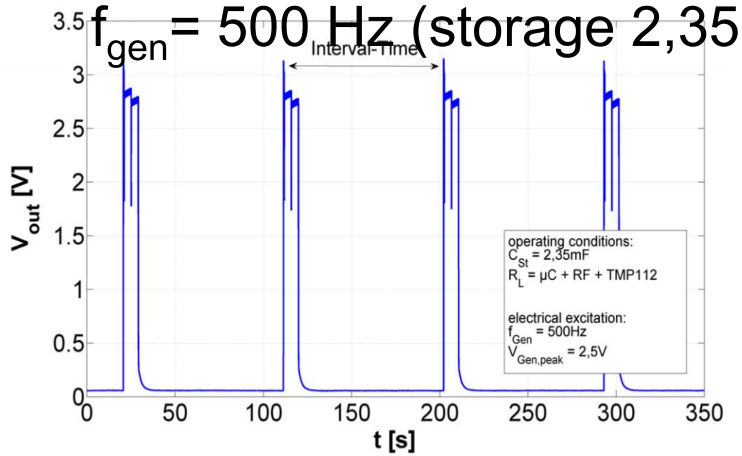


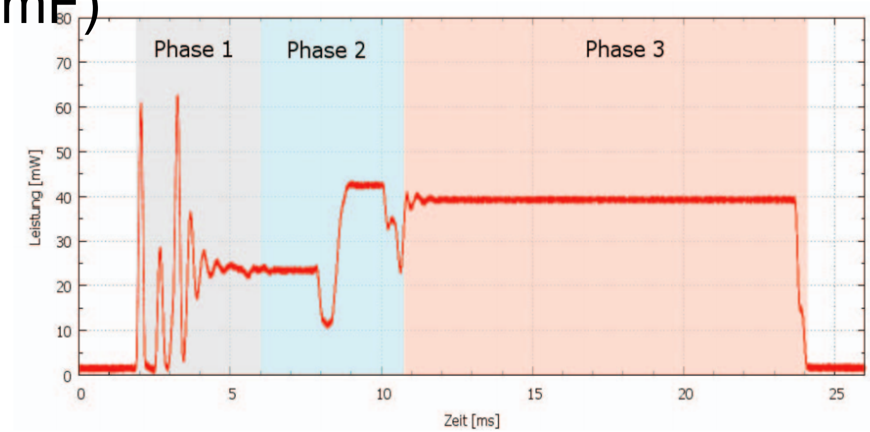


(a) front view

Zimmermann et al., 2012 – 9th International Multi-Conference on Systems, Signals and Devices

f_{gen} = 500 Hz (storage 2,35 mF)





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Piezoelectric Vibrational Energy Harvesters

Requirements:

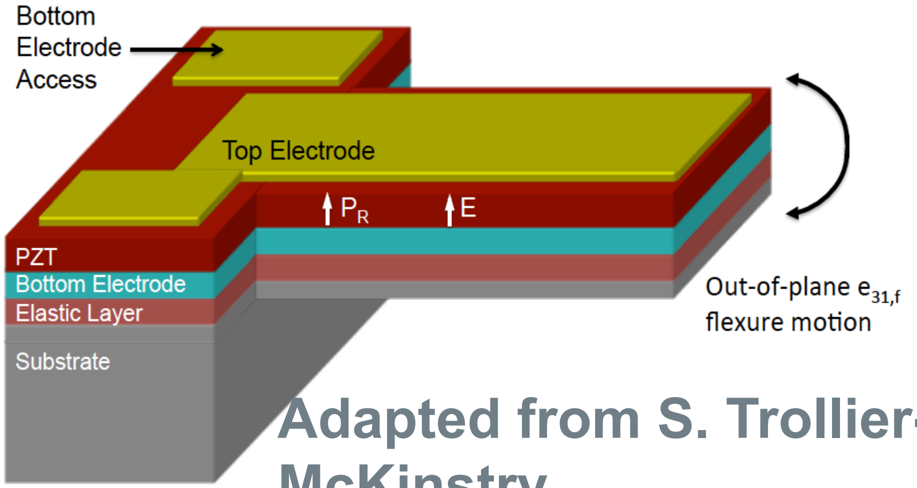
- Compatible with available and variable vibrations in the range of 100 - 500 Hz
- Si compatible technology for direct integration with MEMS sensors or metalic substrates (flexibility, higher thermal expansion)
- High operational temperatures (up to 600 °C)
- Continuous supply of energy for sensor
- Size < 1 cm^3

At present 100s $\mu W/cm^2/g^2$ < necessary power for wireless sensors (harvester with 0.1g of mass excited at 10Hz can provide a maximal power of about 100μW if its volume is 1cm^3)

Possible approaches:

- Advanced materials (FOM = 10-20 GJ/m^3)
- Optimized design of harvesters (max deflexion, degrees of freedom)
- Electronics (CMT – x 6 efficiency; multifrequency- 25%)
- Hybrid energy harvesting (ex. vibrational/ thermal – increase 10%)
- Efficient and long-life energy storage

Piezoelectric Materials for Energy Harvesters



Adapted from S. Trollier-McKinstry

Kamel et al., J. Micromech. Microeng., [20] (2010) 105023

Figure of Merit (FoM)

$$P_{\max} = \frac{1}{4}\left(\frac{e_{31}^{2}}{\varepsilon_o \varepsilon_{33}^{T}}\right)\left(\frac{1-\nu_b}{Y_b}\right)\left(\frac{m}{\omega_o}\right) Q_{tot}^{2} a_o^{2}$$

CHEMICAL REVIEWS
Review
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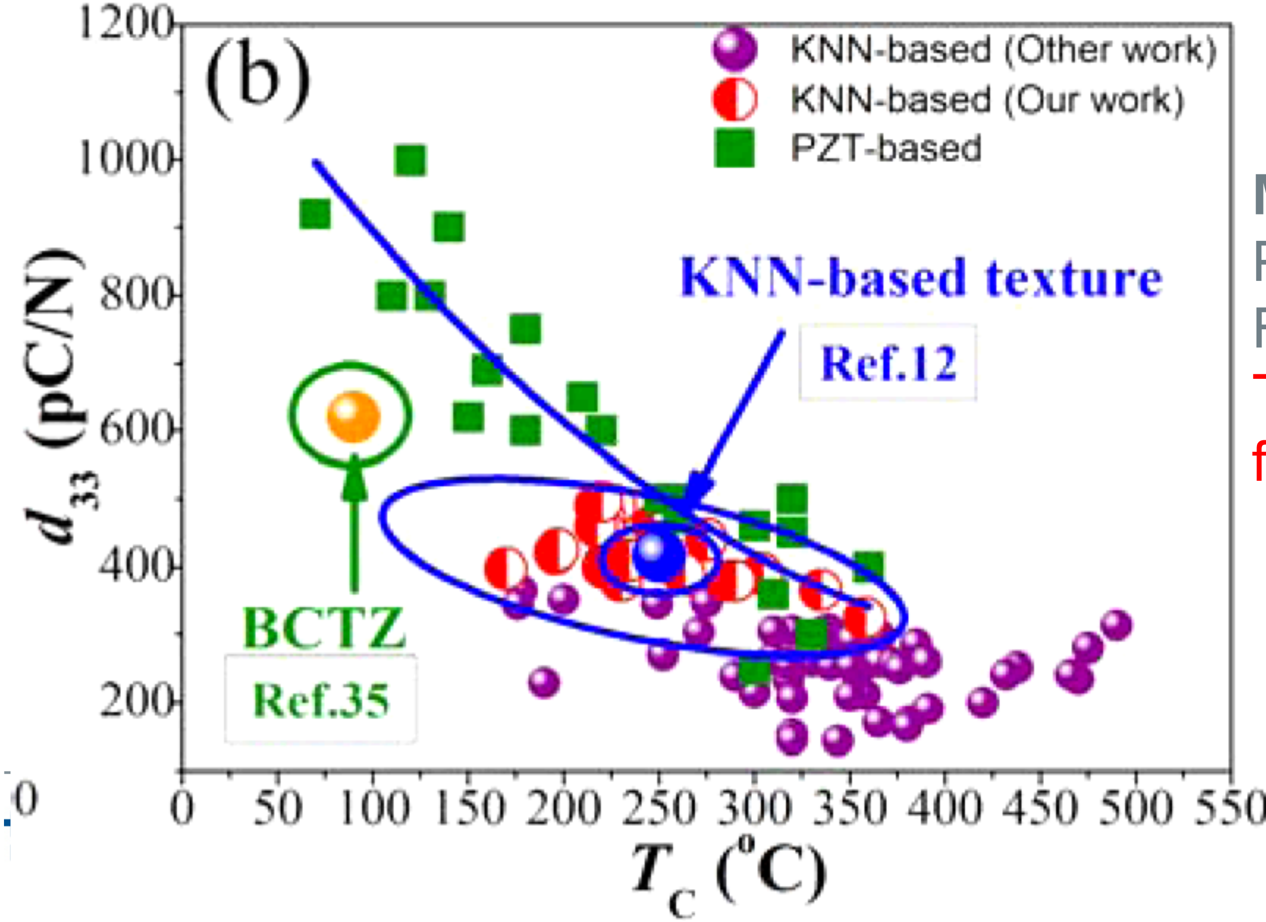
Potassium–Sodium Niobate Lead-Free Piezoelectric Materials: Past, Present, and Future of Phase Boundaries

Jiagang Wu,* Dingquan Xiao, and Jianguo Zhu



Material of choise:

$PbZr_{1-x}Ti_xO_3$ (high K^2 & Figure of Merit (FoM))

To be replaced by lead-free materials !

Piezoelectric Materials for Energy Harvesters

Material of choise: $PbZr_{1-x}Ti_xO_3$ (high K^2 & Figure of Merit (FoM))

To be replaced by lead-free materials !

	AlN	ZnO	$BaTiO_3$	Hard PZT	Soft PZT	PMN-PT	$LiNbO_3$	PVDF
Const. Stress rel. Perm. ε^T_{33}	11.9	11	1200	1300	3400	8200	28.7	7.6
d_{33}, pC/N	5	12.4	149	289	593	2820	6	-33
d_{31}, pC/N	-2	-5.0	-58	-123	-274	-1330	-1	21
d_{15}, PC/N	3.6	-8.3	242	495	741	146	69	-27
Mechanical quality factor	2490	1770	400	500	65	43-2050	10^4	3-10

Kamel et al., J. Micromech. Microeng., [20] (2010) 105023

$$P_{\max} = \frac{1}{4}\left(\frac{e_{31}^2}{\varepsilon_o \varepsilon_{33}^T}\right)\left(\frac{1-\nu_b}{Y_b}\right)\left(\frac{m}{\omega_o}\right) \mathbf{Q}_{tot}^2 a_o^2$$

Bowen et al.Energy Environ. Sci., 2014, 7, 25

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Piezoelectric Materials for Energy Harvesters

APPLIED PHYSICS LETTERS **102**, 021915 (2013)

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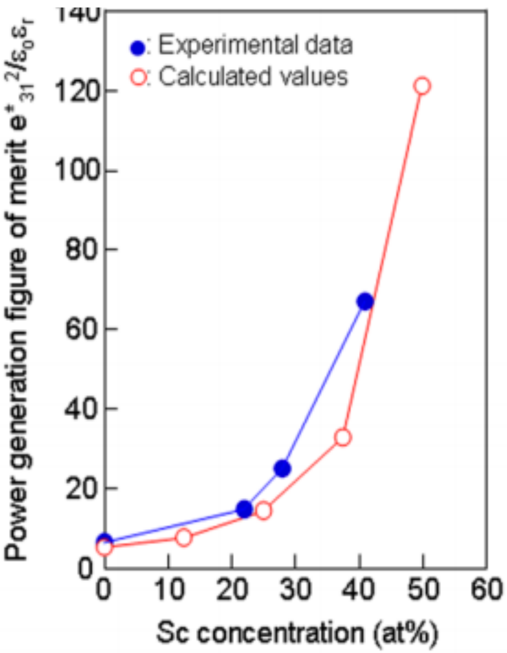
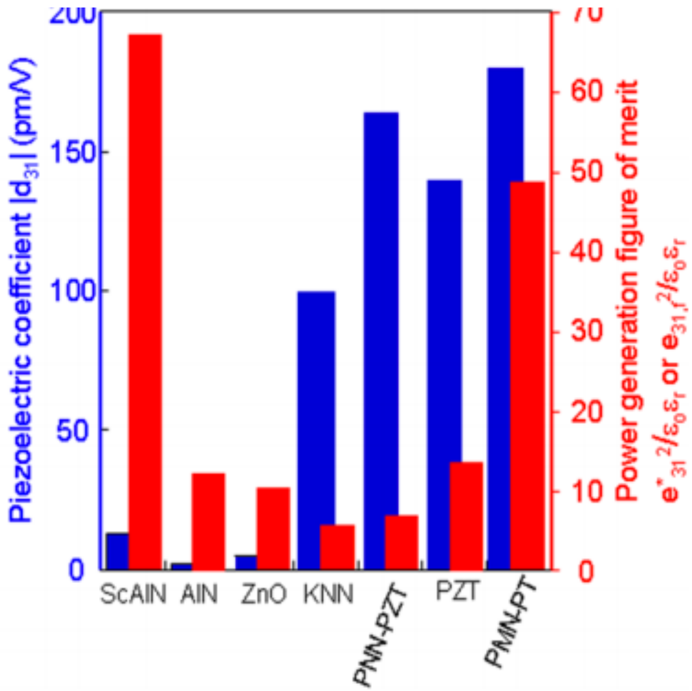
Influence of scandium concentration on power generation figure of merit of scandium aluminum nitride thin films

Morito Akiyama,[1] Keiichi Umeda,[2] Atsushi Honda,[2] and Toshimi Nagase[1]
[1]*Measurement Solution Research Center, National Institute of Advanced Industrials Science and Technology, Tosu, Saga 841-0052, Japan*
[2]*Murata Manufacturing Co., Ltd., Nagaokakyo, Kyoto 617-8555, Japan*





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HIGH PERFORMANCE NONLINEAR MICRO ENERGY HARVESTER INTEGRATED WITH (K,Na)NbO_3/Si COMPOSITE QUAD-CANTILEVER

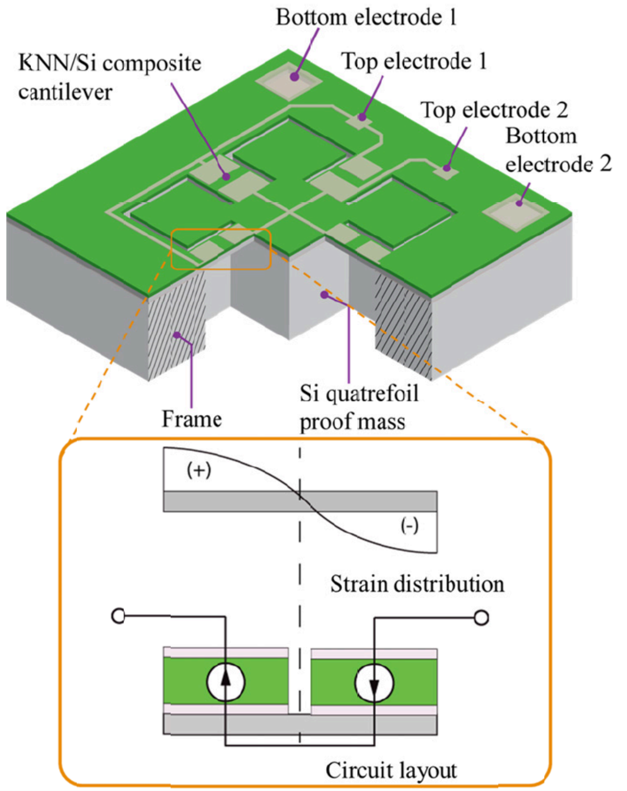
Le Van Minh, Motoaki Hara, and Hiroki Kuwano
Tohoku University, Sendai, JAPAN

IEEE Int. Conf. MEMS, 397 (2014)

Table 1: Geometric parameters of the proposed energy harvester

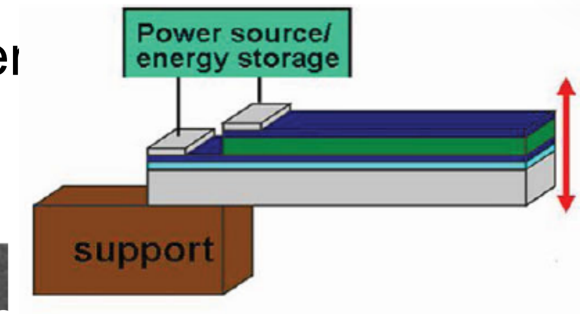
Length	1500 µm
Width	500 µm
KNN thickness	2 µm
Si thickness	10 µm
Total mass	90 µg
k_0	0.2146 µN/µm
k_1	0.5786×10^{-3} µN/µm^3

The harvester showed wide bandwidth of 253 Hz (fractional bandwidth: 12.9%) at acceleration of 6 m/s^2. The power density of 1623 µW/cm^3 was achieved at the same acceleration. It is the highest value among the wide bandwidth piezoelectric energy harvesters.

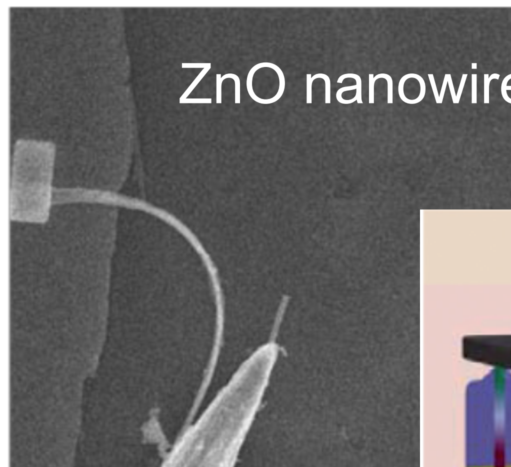


Integrated devices/ nanostructures

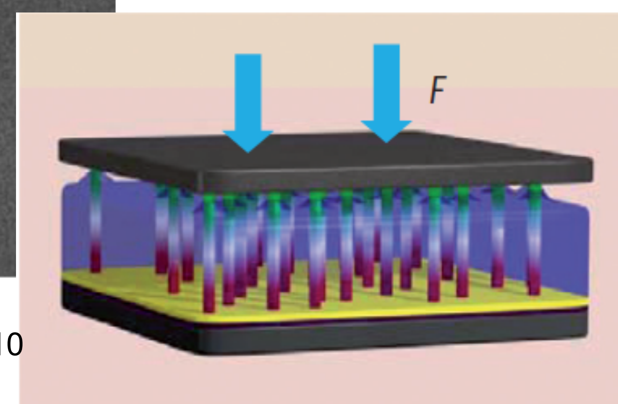
- Increased elastic compliance
- Elasticity without cracking
- Able to convert several different mechanical en
- Fatigue
- Up-scalling & cost
- Environmental issues
- Non-resonant mode

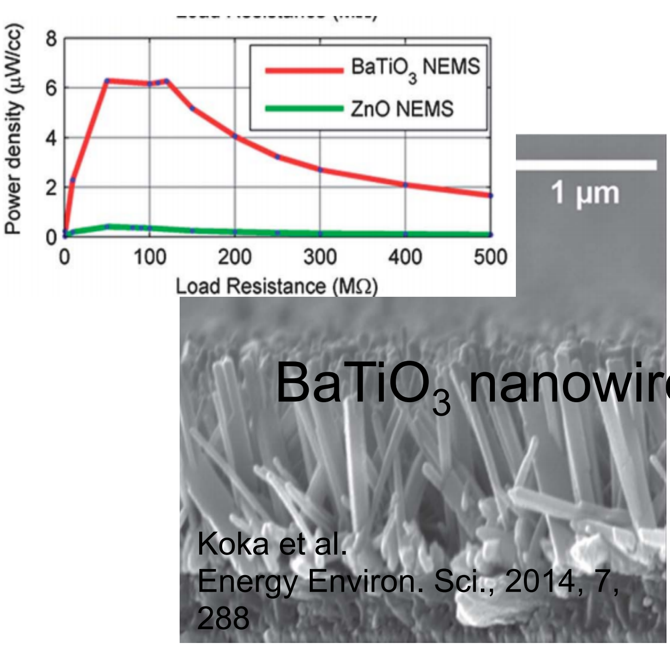


Kursumovic et al.
Nature Adv. Func. Mat. 2013



Xu et al.
Nature Nanotechnology 2010





Koka et al.
Energy Environ. Sci., 2014, 7, 288

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Advanced materials for energy harvesters

Optimisation of growth processes and study of physical properties of different potential **lead-free materials, enabling hybrid EH**:

i. $LiNbO_3$ (< 1150 °C)

ii. $BiFeO_3$ (< 500 °C)

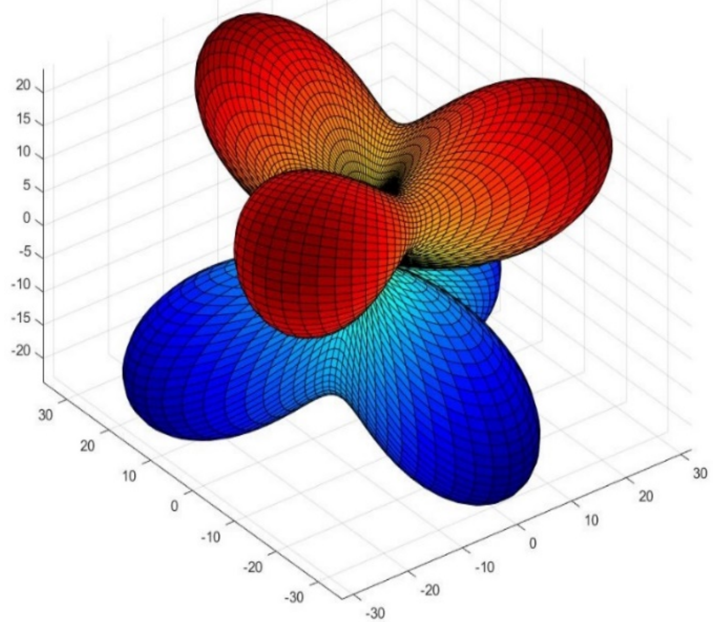
iii. $K_{1-x}Na_xNbO_3$ (<200-400 °C)

- Similar performance to PZT
- Piezoelectric-Pyroelectric-Photovoltaic
- High-temperature piezoelectricity

However, the application of these materials in PiViEHs is still very little studied and considerable efforts have to be done towards their integration to the conventional processing of MEMS.

Properties of $LiNbO_3$

Piezoelectric tensor of $LiNbO_3$



Characteristics:

- Lead free
- High Curie temperature
- High quality crystal (low price)

- Piezoelectric coefficients (pC/N)

$$d = \begin{pmatrix} 0 & 0 & 0 & 0 & \boxed{68} & \boxed{-42} \\ -21 & 21 & 0 & \boxed{68} & 0 & 0 \\ -1 & -1 & 6{,}2 & 0 & 0 & 0 \end{pmatrix}$$

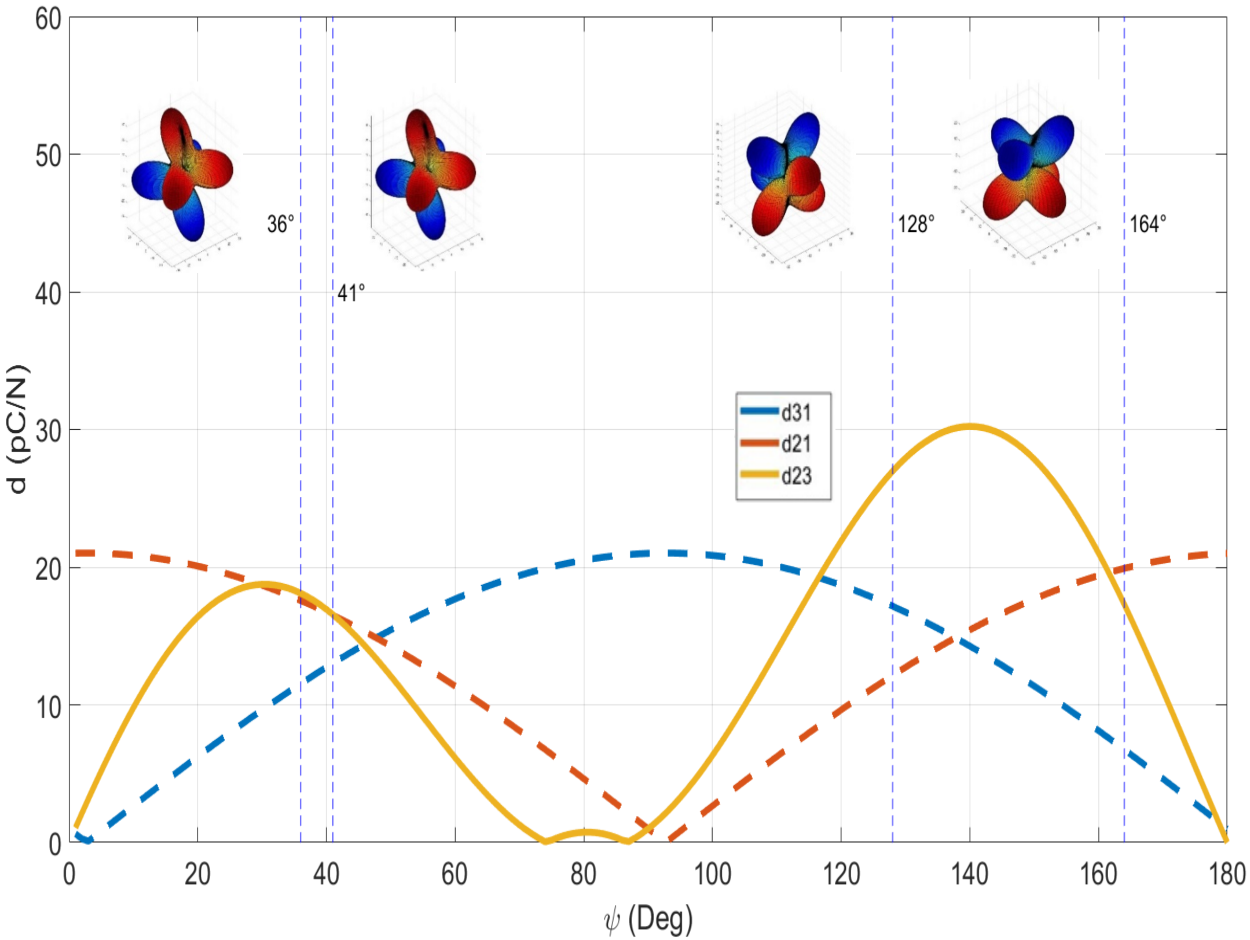
Highlighted value are shear (d_{15}) coefficients.

- Electromechanical coupling factor: $k_{31}^2 = \dfrac{d_{31}^2}{s_{11}^E \varepsilon_{33}^T}$

What is the best orientation?

- Available Y cuts: 0°, 36°, 41°, 128°, 168°
- Other cuts: Z cut, X cut
- Thin Film: Y 33° epitaxial growth

Piezoelectric tensor orientation study



Vertical dashed lines represent available Y cuts.

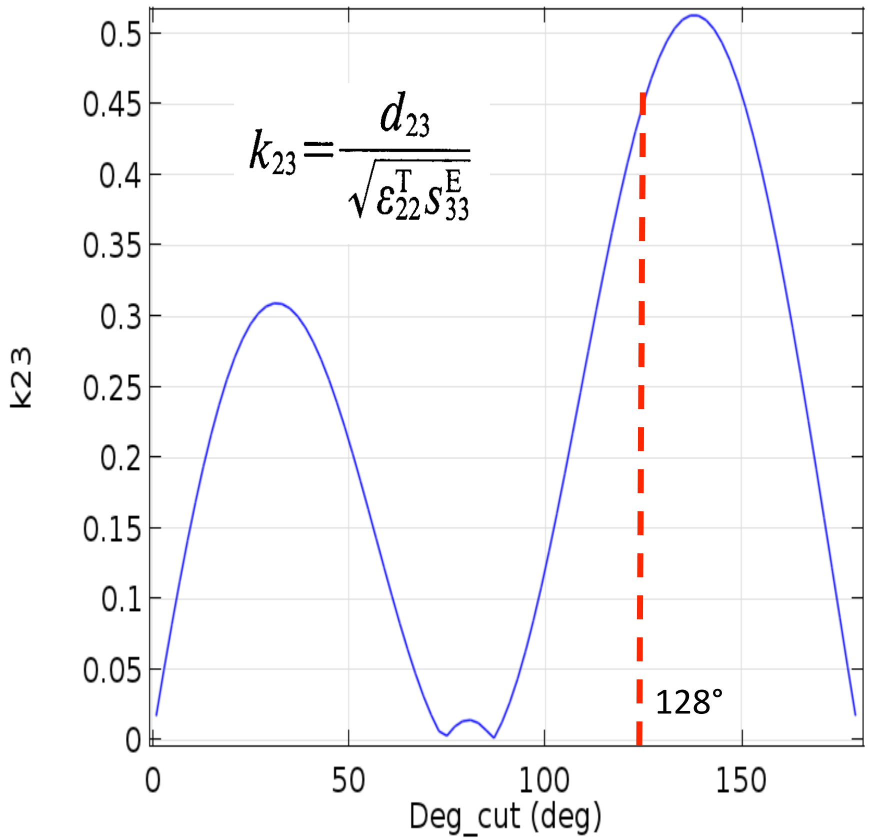
Maximum values:

- d_{31} 21 pC/N at angles of **87°** .
- d_{21} is 21 pC/N **168°**.
- d_{23} is 27 pC/N at **128°**.

Rotated Y-cut plates with the length along the Z axis are suitable for bending structures.

Electromechanical coupling study

Electromechanical coupling for d_{23} mode



Lithium niobate Y cut 128° $k_{23}=0.45$

Material	d31 (pC/N)	e33	k31
PZT-5H	-270	1400	0.44
AlN	-2	9	0.13
ScAlN	14	16	0.52
PVDF	13	7.3	0.12

Table of piezoelectric transducers

Low dielectric constant & high d_{15}

$LiNbO_3$ & Vibrational Energy Harvesting

140 ° rotated Y-cut $LiNbO_3$ single crystal

Funasaka et al., *IEEE Ultrasonic Symposium 956 (1998)*

Table 1 Coupling factor, Q, and generation efficiency (η_p) of PZT and $LiNbO_3$ generators

			PZT	$LiNbO_3$
fr	free	(kHz)	10.2	17.8
k^2	free	(%)	6.0	12.5
Q	free	(±0.5V)	580	2640
fr	fix	(kHz)	2.71	4.75
k^2	fix	(%)	4.3	8.0
Q	fix	(±0.5V)	330	1040
Q	impact	(±10V)	200	229
η_p		(%)	65	78

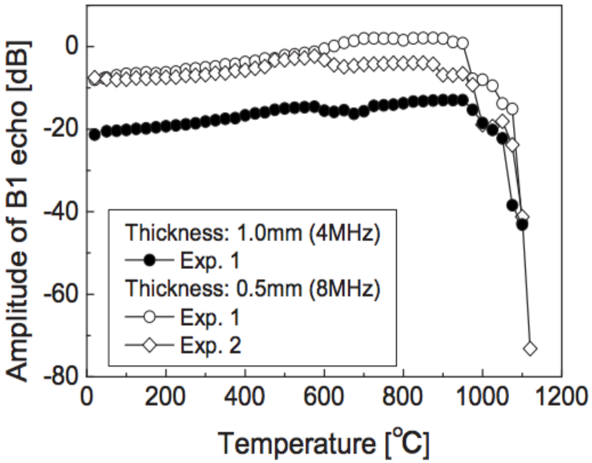


FIG. 3. Comparison of the amplitude of B1 echo from the substrate.

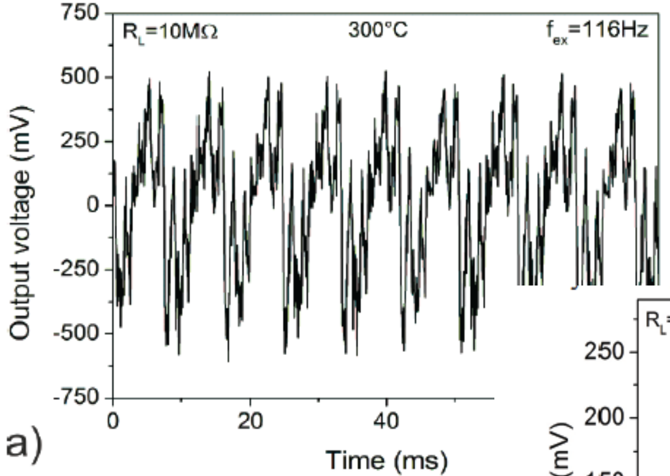
High-temperature energy harvesting

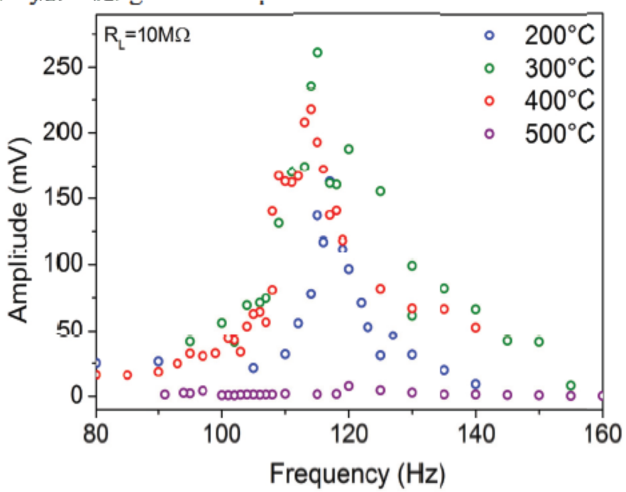
-transducers based on $LiNbO_3$ crystals operating up to 1000°C (Baba et al. Appl. Phys. Lett.,2010, 97, 232901)

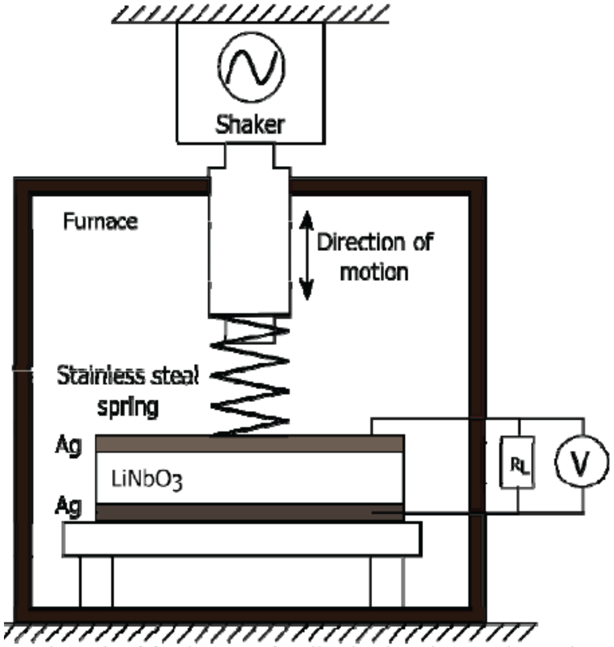
$LiNbO_3$ & Vibrational Energy Harvesting

High-temperature energy harvesting
Y-cut $LiNbO_3$ single crystal

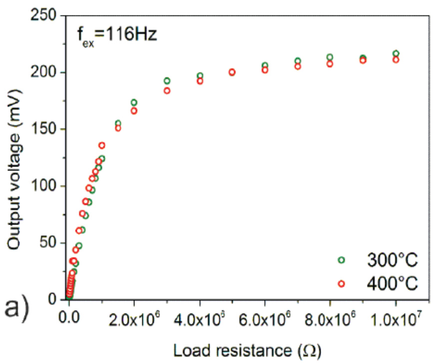
Boulbar et al., ISAF/ISIF/PFM proceedings, 292 (2015)



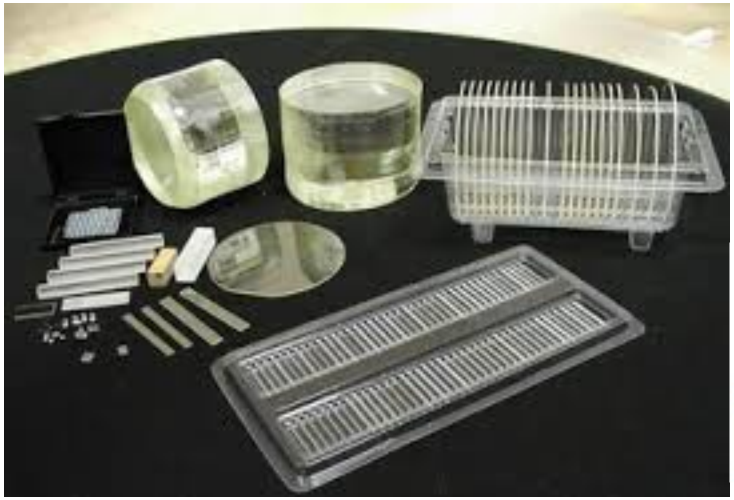




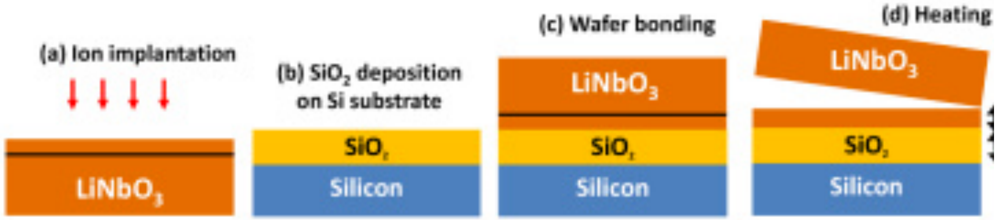
3: Schematic of the rig set-up for vibration-based energy harvesting at high temperature



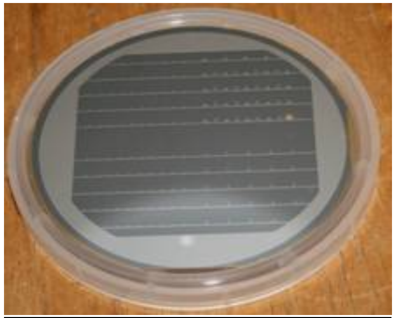
Lead –free piezoelectrics: $LiNbO_3$ & $LiTaO_3$



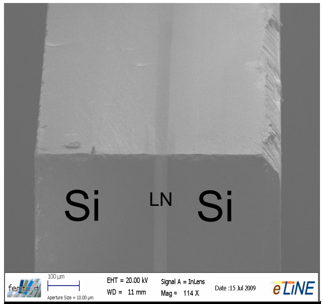
Smart-cut of $LiNbO_3$ films (expensive)



- $LiNbO_3$ and $LiTaO_3$ single-crystal films wafer bonded to the Si or metal for demonstrators: free choice of substrate and film orientation



$LiNbO_3$/Si



Si / $LiNbO_3$ / Si
Gold bonding

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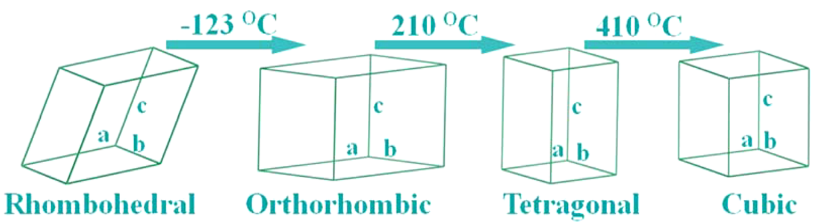
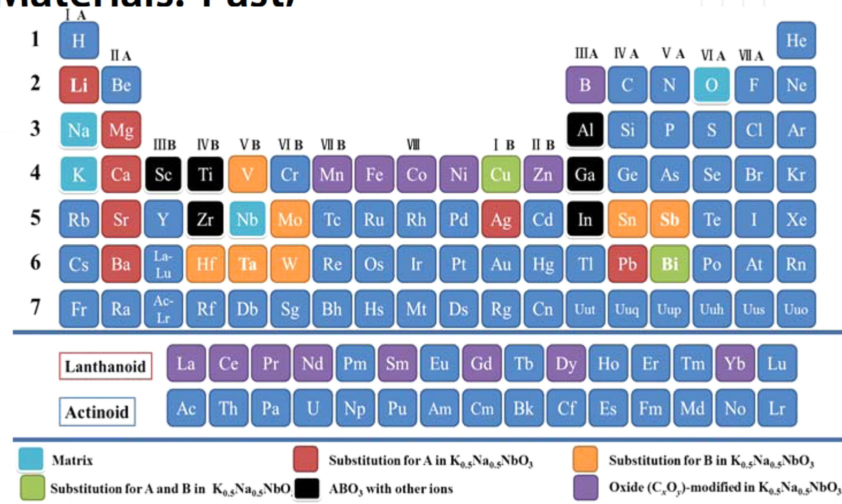
Potassium–Sodium Niobate Lead-Free Piezoelectric Materials: Past, Present, and Future of Phase Boundaries

Jiagang Wu,* Dingquan Xiao, and Jianguo Zhu





4.5.7. Thin Films. Since the increase in interest in the piezoelectric activity of KNN-based ceramics, efforts have been made to prepare corresponding thin films due to their potential applications for microdevices such as microelectromechanical systems.[552−590] However, two main issues still hinder the fabrication of high-quality KNN-based films: (i) composition deviations from stoichiometry and (ii) the loss of alkali oxides during preparation.[553,573,589,590] Table 20 lists the preparation

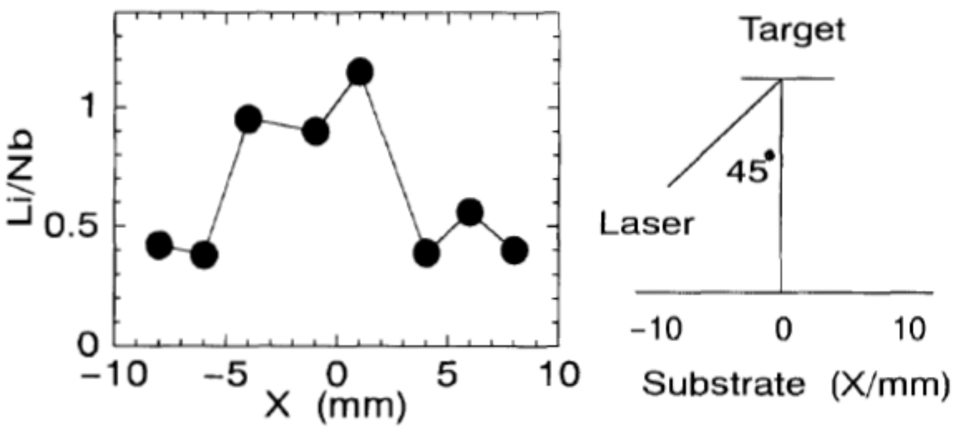
Control of alkali metal oxide composition

Li_2O, K_2O, Na_2O are highly volatile:

Deposition under vaccum conditions:

Composition depends on film thickness !!
Composition needs to be optmized for each thickness !!

Physical methods (PLD, magnetron sputtering, etc.):
Very difficult task at high deposition temperatures and very low pressures



Y. Shibata, Proc. IEEE Ultrason. Symp. ***1996***

Liquid phase epitaxy:
C-$LiNbO_3$ thick films on C-$LiTaO_3$

ALD: %Li independent on thickness, crystallization of amorphous films may result in worse epitaxial quality, adapted for very thin films

MOCVD:
fine tuning of composition, high epitaxial quality

Atmospheric pressure MOCVD: composition independent on thickness but worse epitaxial quality

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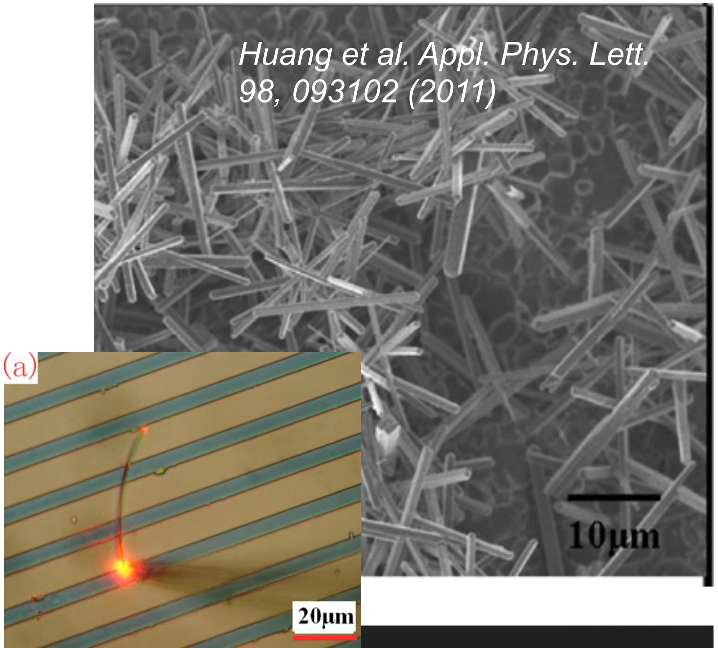
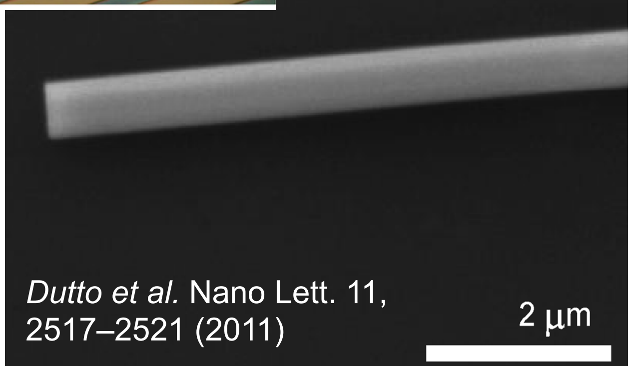
LiNbO$_3$ nanostructures

Nanophotonic devices

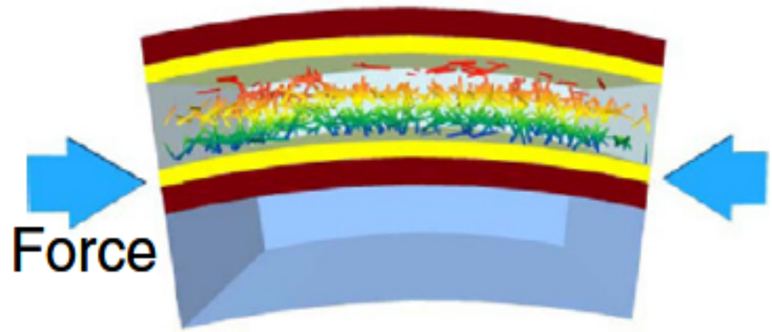


Huang et al. Appl. Phys. Lett. 98, 093102 (2011)

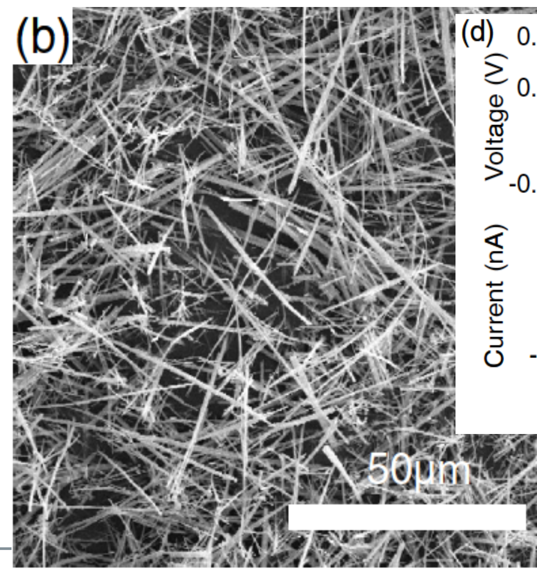


Dutto et al. Nano Lett. 11, 2517–2521 (2011)

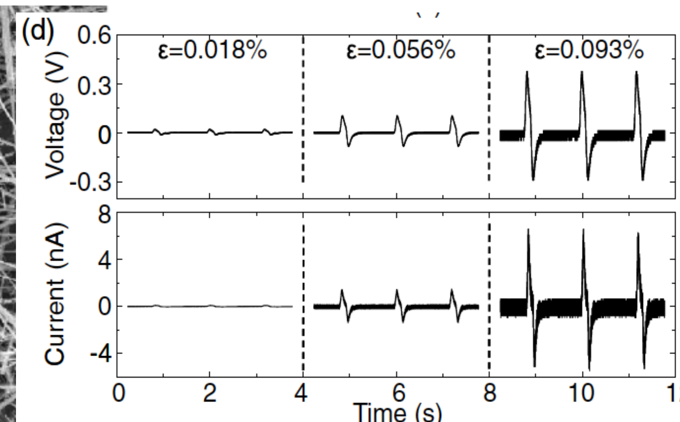
Nano- or micro-generators



Yun et al. Nanoscale Research Letters 2014, 9:4



Hydrothermal synthesis



Similar performance to that of ZnO nanowires

Hybrid energy harvesting

In the working environment of the sensor, the available energy source may vary from time to time or from location to location (parked car, night, dark places, ect.)

Hybrid energy harvesters - increased efficiency and ability to scavenge different types of energy and to supply more stable power supply for sensor nodes.

Hybrid energy harvesters started to be studied these last years:

- Magnetic-inertial (bulk) piezoelectric harvester
- Electrostatic-piezoelectric harvesters,
- Solar-vibration piezoelectric harvesters
- Thermal-inertial harvesters **(reduced efficiency)**
- Thermal-solar harvesters
- Etc.

In general, the above cited **hybrid harvesters were realized by the integration of two or several sensitive elements/transducers**. Consequently, the final volume of the structures and the conditioning electronics also increased with the number of the elements

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Integrated smart systems

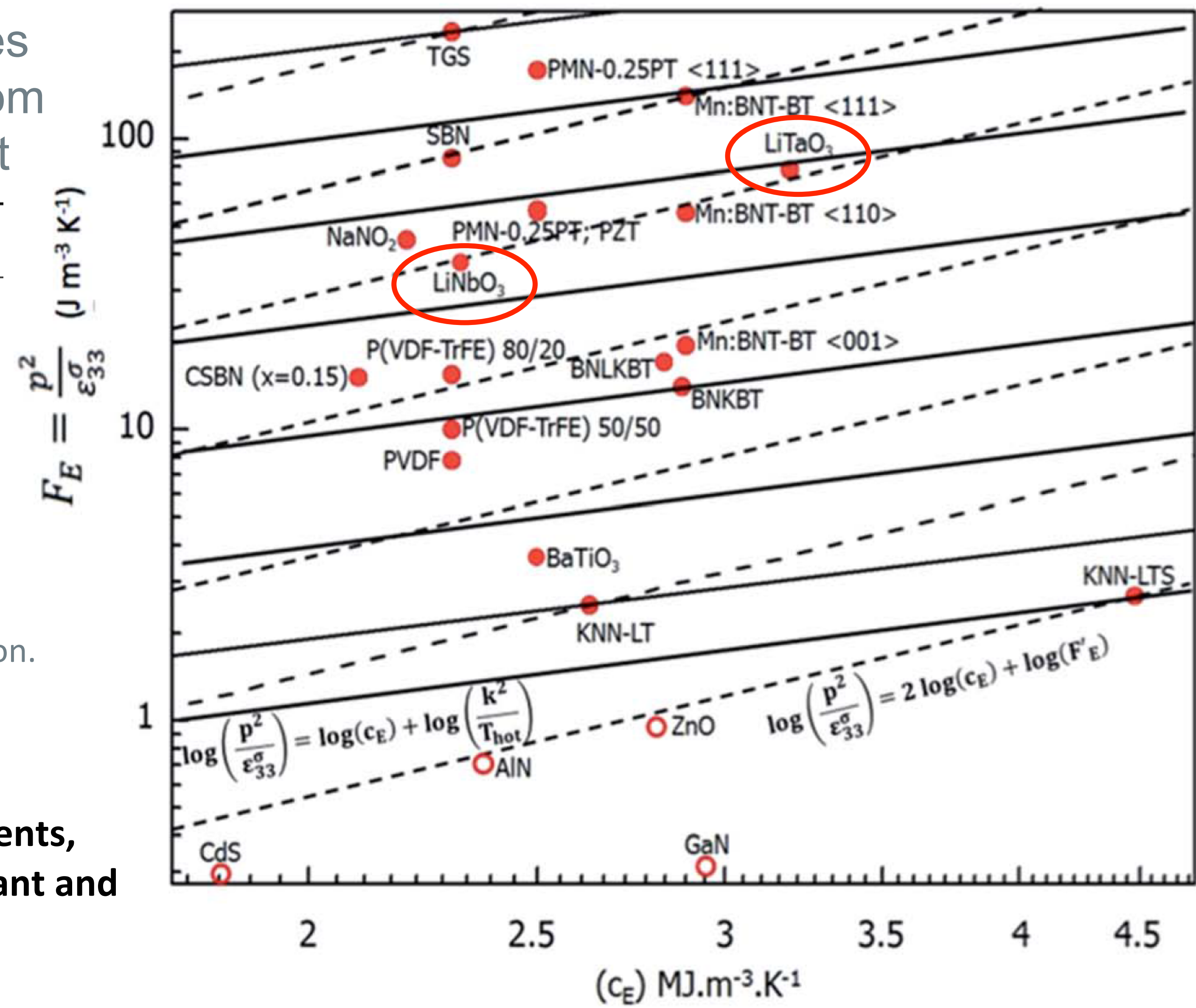
Materials for Pyro-Piezo EHs

Additional charges can be gained from pyroelectric effect

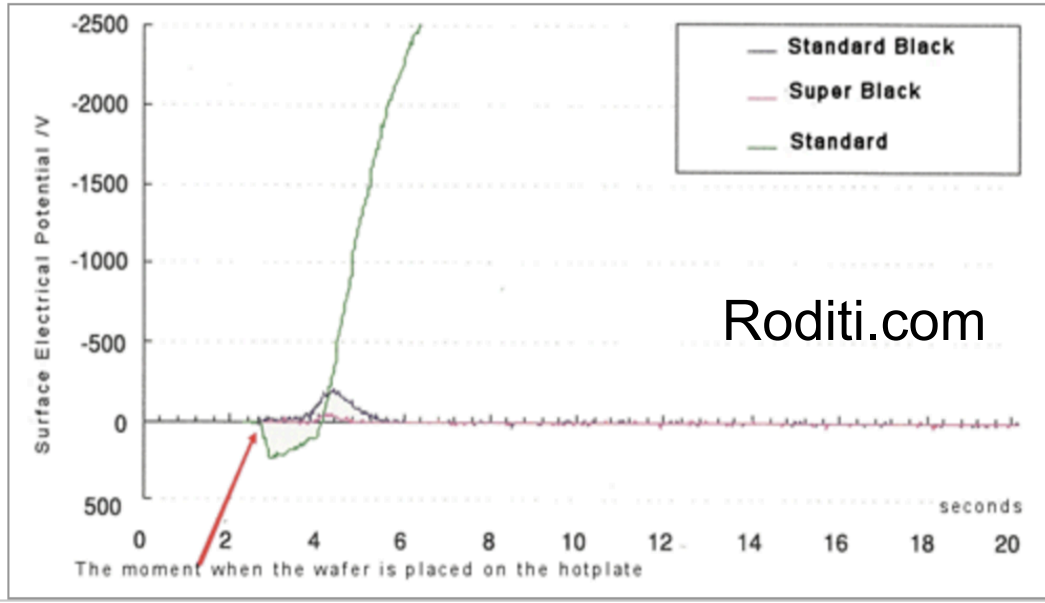
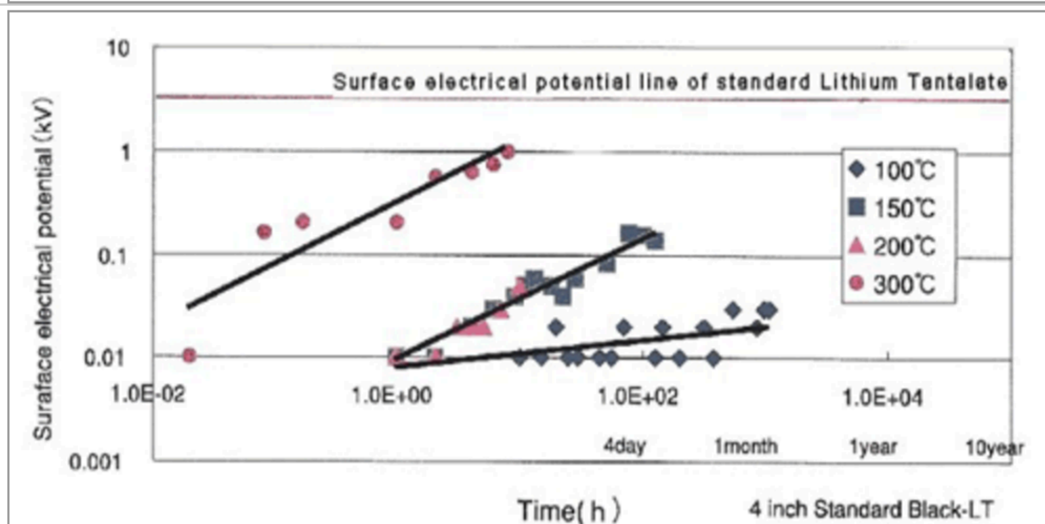
Pyroelectric
$Q = pA\Delta T$
$i = pA\dfrac{\Delta T}{\Delta t}$
$V = \dfrac{p}{\varepsilon_{33}^{T}} h\Delta T$
$E = \dfrac{1}{2}\dfrac{p^2}{\varepsilon_{33}^{T}} Ah(\Delta T)^2$

Bowen et al., Energy Environ. Sci., 2014, 7, 3836

- Materials with **high pyroelectric coefficients, low dielectric constant and fast heat transfer**



Materials for Pyro-Piezo EHs

Comparison of electrical potential between black lithium tantalate and standard lithium tantalate when placed on a hotplate at 95Â°C	
Black Lithium Tantalate - Endurance Test against High Temperature	

Very few reports on the energy harvesters based on $LiNbO_3$-$LiTaO_3$

Dynamic & nonlinear modelling, interval techniques, control theory

Designer vibrational energy harvesters and actuators with increased performance, selfsensing & devices with multi-degrees of freedom

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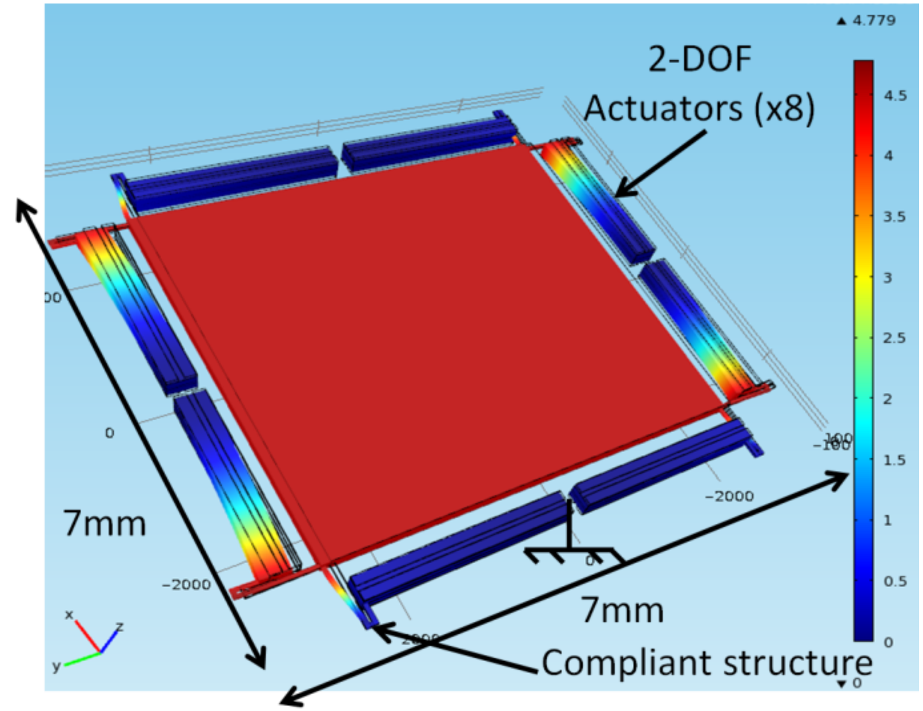
Design, modeling and cahracterization of hybrid energy harvesters

Design & mechanical modelling using automatics and control tools of:

i. Optimised **multi-frequency and multidegrees of freedom** (able to harvest vibrations with different motion vector and of different origins) PiViEH systems permitting to maximise charge extraction and to increase the electrical damping at the mechanical resonant frequency;

ii. Mechanical structure of hybrid thermal-vibrational EHs enabling addition of pyroelectric, thermal expansion and piezoelectric effects and able to harvest efficiently up to 600°C;

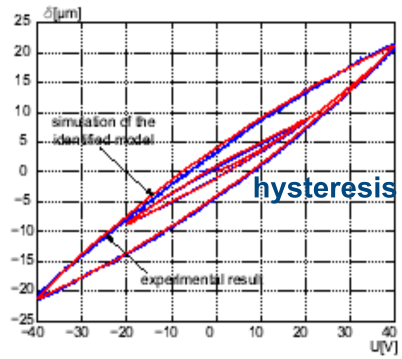
<u>Optimal designed 2-DOF piezoelectric structure (M. Rakotondrabe et al, UFC)</u>

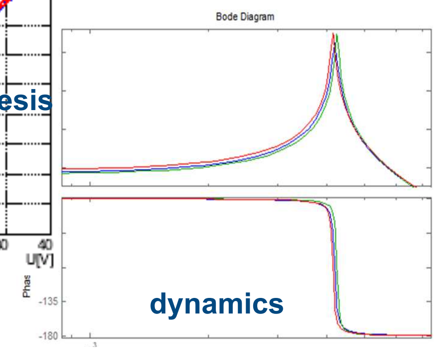


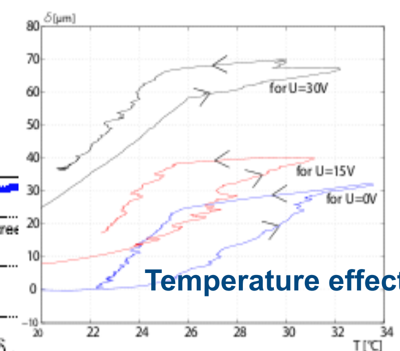
Nonlinear and dynamic modeling of piezoelectric devices

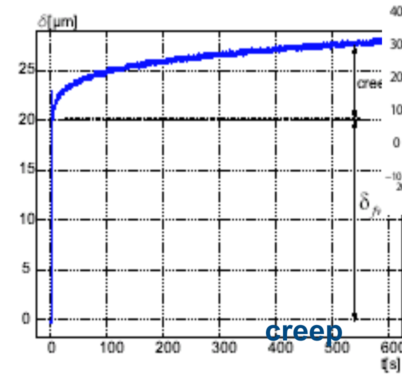
$$y = D_{dyn}(s)\Gamma\left(U\right) + C(s)U + f\left(\Delta T\right) + g\left(F, M\right)$$

hysteresis and dynamics | **creep** | **temperature effect** | **mechanical effect**









The models account for:

-the hysteresis nonlinearity *(Prandtl-Ishlinskii, Bouc-Wen, Preisach and quadrilateral approach),*

-the creep phenomenon *(linear approximation),*

-the dynamics *(high orders precise model with multiple resonance),*

-the mechanical effect *(for instance ambiant vibration).*

The models are interesting and even essential for

-optimal design of actuators,

-robust and optimal controllers synthesis,

-optimal design of sensors,

-design of energy harvesters.

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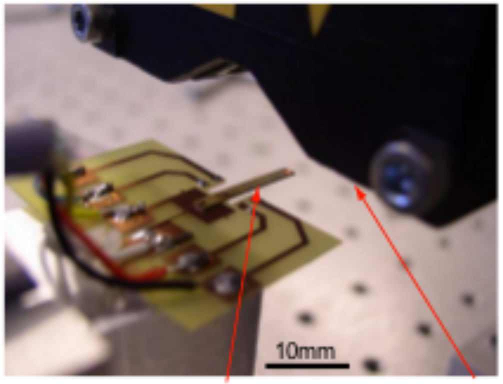
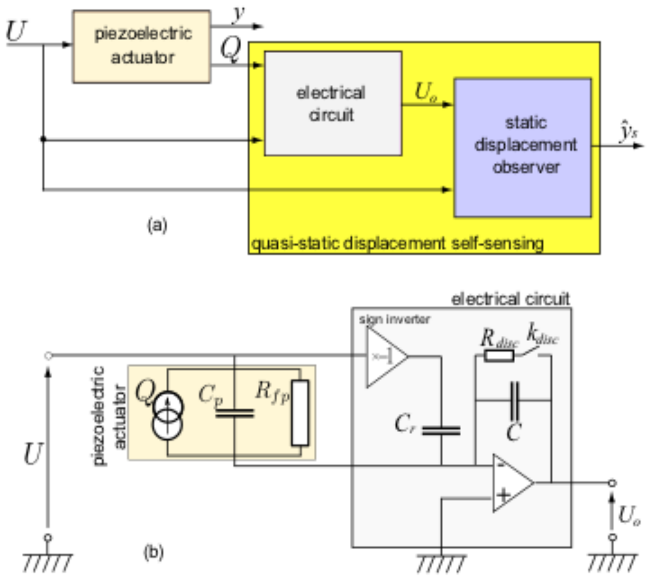
Nonlinear and dynamic modeling of piezoelectric devices

Self-sensing: utilization of the same material as simultaneously sensor and actuator/harvester:
-no external sensors required,
-high level of packageability.



piezoelectric cantilever displacement sensor



Ongoing works:

-possibility of utilization of the self-sensing in a piezoelectrically feedbacked energy harvester in order to maintain the system oscillating at resonant frequency.

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Interval techniques for robust and optimal design of piezoelectric devices

Piezoelectric models
with physical and geometrical parameters

Required performances
-minimal output amplitudes
-resonant frequencies
-Q-factor
-...

Performances inclusion theorem [Rakotondrabe, ACC2011]

Given two stable interval transfers $[G_1](s,[a_1],[b_1])$ **and** $[G_2](s,[a_2],[b_2])$ **having the same structure.**

If $\begin{cases}[a_1]\subseteq[a_2]\\ [b_1]\subseteq[b_2]\end{cases}$ **then** $[G_1](s,[a_1],[b_1])\subseteq[G_2](s,[a_2],[b_2])$ **and therefore the performance**

of $[G_1](s,[a_1],[b_1])$ **are bounded by those of** $[G_2](s,[a_2],[b_2])$

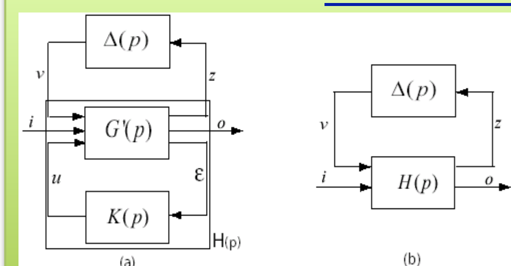
-Optimized piezoelectric devices
(actuators, sensors, harvesters)

Control theory (feedback techniques) for optimal design of piezoelectric devices

Piezoelectric models
with physical and geometrical parameters

Required performances
-minimal output amplitudes
-resonant frequencies
-Q-factor
-…

Feedback controllers synthesis



- **-small gain theorem**
- **-Passivity, dissipatiity techniques**
- **-Lyapunov techniques**
- **-Port-Hamiltonian systems approach**

$$\dot{x}(t) = J\mathcal{H}x(t) + Bu(t)$$
$$y(t) = B^{*}\mathcal{H}x(t)$$

-Optimized piezoelectric devices
(actuators, sensors, harvesters)

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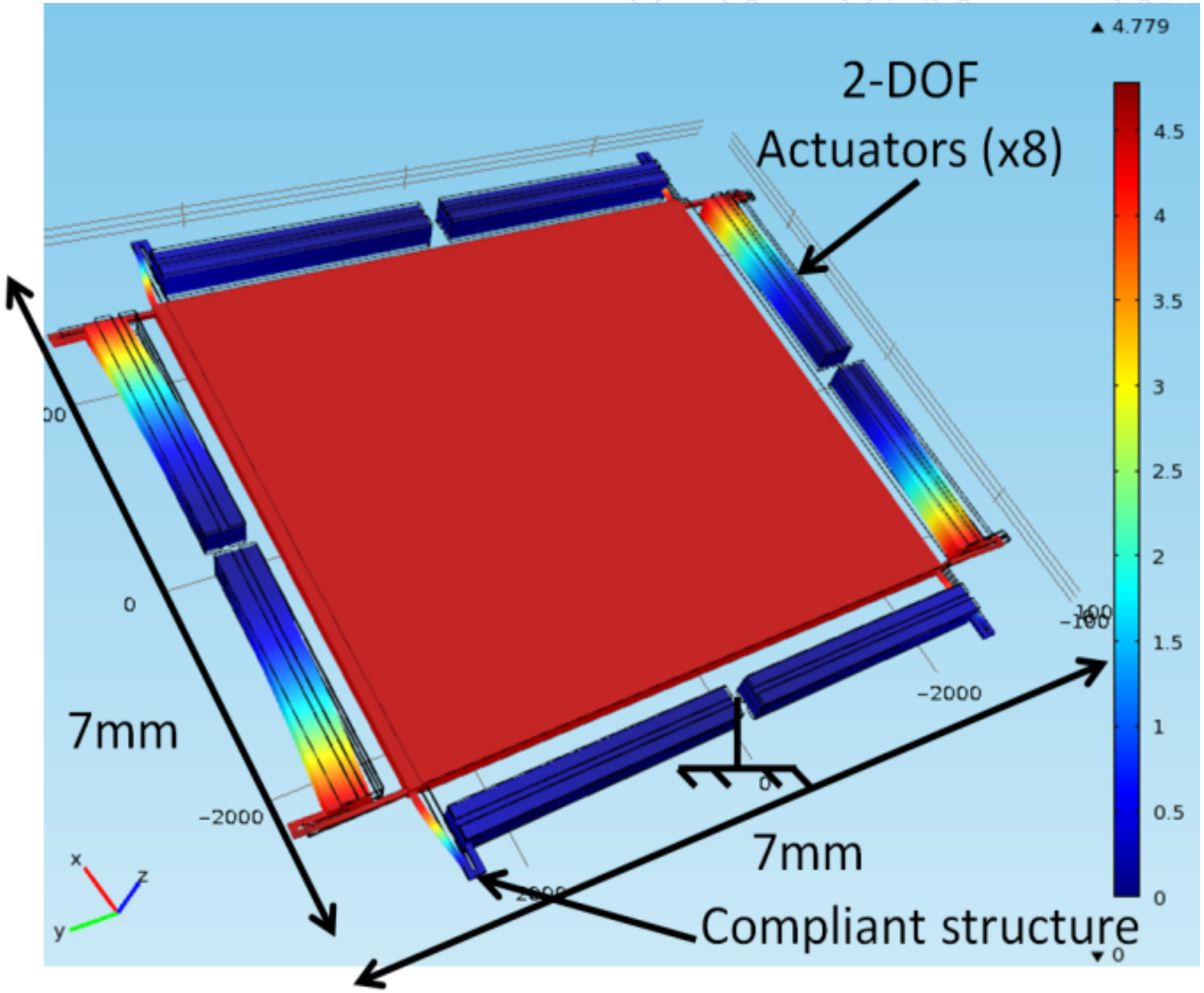
High performances multi-degrees of freedom (DOF) piezoelectric devices

Optimal designed
2-DOF piezoelectric structure



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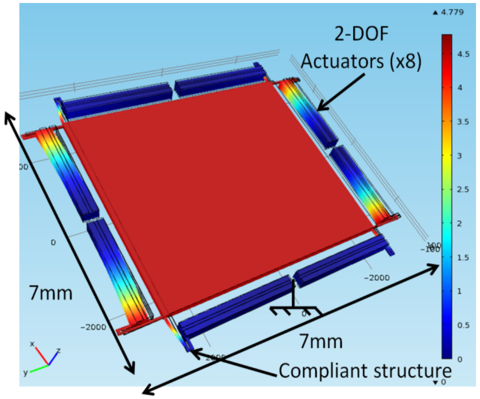
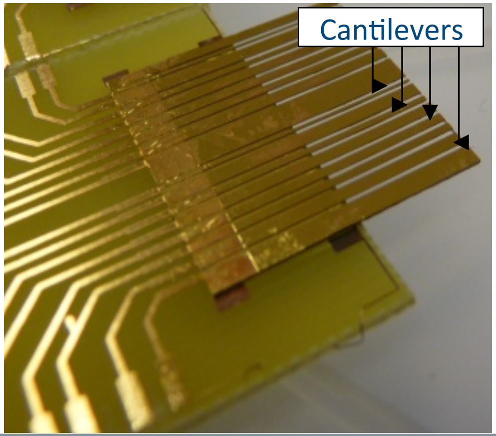
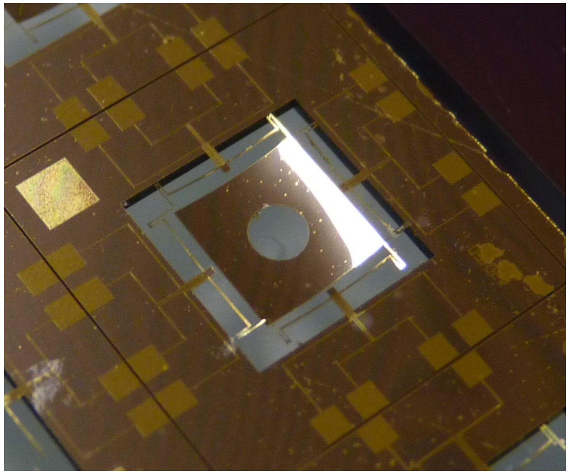
High performances multi-degrees of freedom (DOF) piezoelectric devices

Optimal designed 2-DOF piezoelectric structure

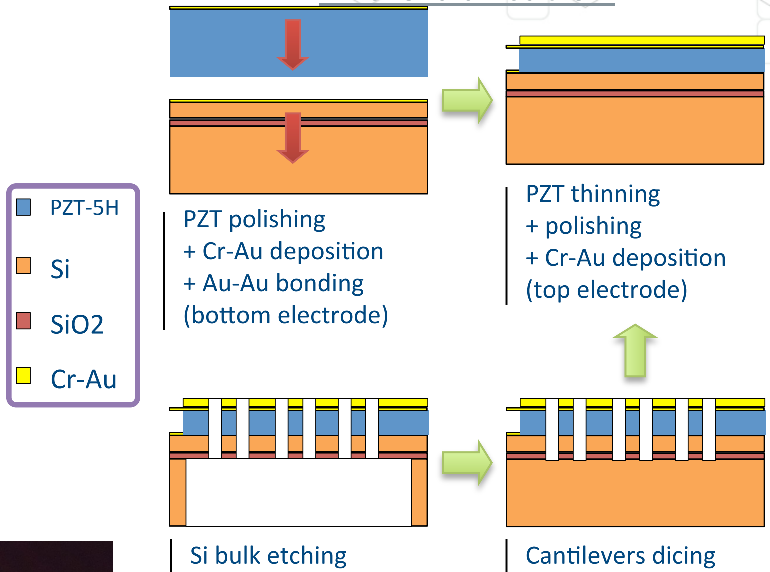


Microfabricated structures





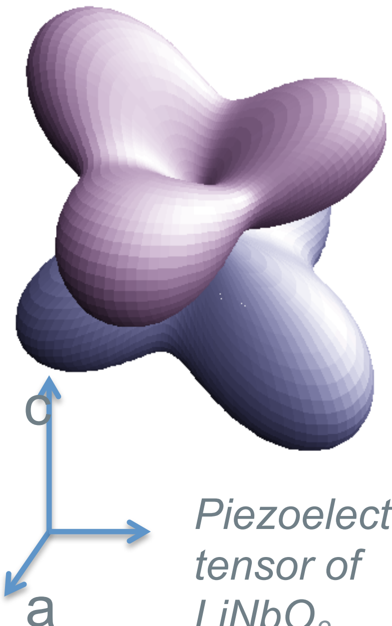
microfabrication



Features

-3 linear DOF (XYZ)
-up to 25µm of displacement
-High bandwidth
-Miniature: 7mmx7mm

Summary



Piezoelectric tensor of $LiNbO_3$

Thank you for your attention!