

## Supporting Information

### A synergistic approach to attain high piezoelectricity in

### **Pb(Ni, Nb)O<sub>3</sub>-Pb(Lu, Nb)O<sub>3</sub>-PbTiO<sub>3</sub> system**

Mengdi Cheng<sup>a</sup>, Yangxi Yan<sup>a</sup>, Zhimin Li<sup>a</sup>, Pangpang Wang<sup>b</sup>, Ri-ichi Murakami<sup>c</sup> and

Dongyan Zhang<sup>a,\*</sup>

<sup>a</sup> *School of Advanced Materials and Nanotechnology, Xidian University, 266 Xinglong*

*Section of Xifeng Road, Xi'an 710126, P.R. China*

<sup>b</sup> *Nanomaterials Group, Institute of System, Information Technologies and*

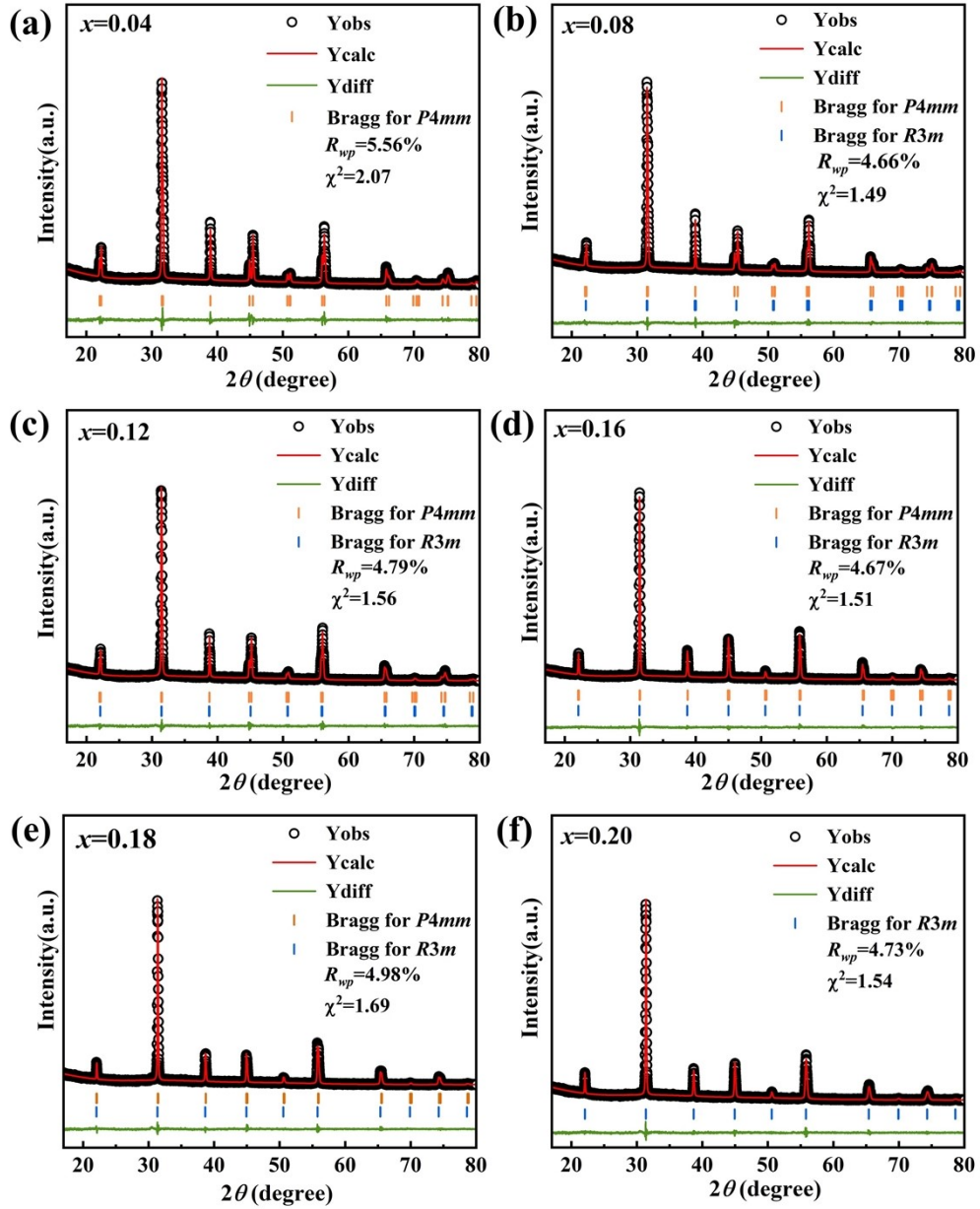
*Nanotechnologies (ISIT), Fukuoka Industry-Academia Symplicity (FiaS), 4-1*

*Kyudaishinmachi, Nishi-ku, Fukuoka 819-0388, Japan*

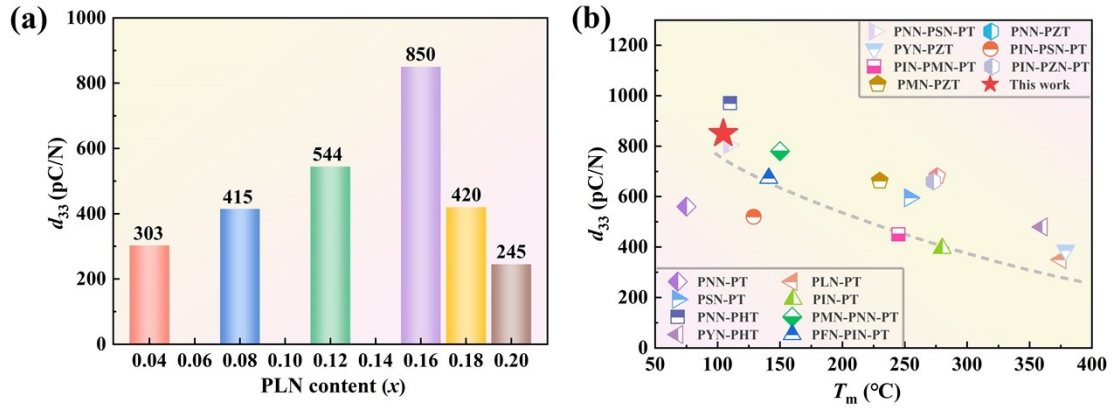
<sup>c</sup> *School of Mechanical Engineering, Chengdu University, No. 2025, Chengluo Avenue, Chengdu*

*610106, P. R. China*

*Corresponding author. E-mail addresses: zhangdongyan@xidian.edu.cn*



**Fig. S1.** Rietveld refinements results of XRD patterns for PNN- $x$ PLN-PT ceramics with  $R3m$  and  $P4mm$  models, (a)  $x = 0.04$ , (b)  $x = 0.08$ , (c)  $x = 0.12$ , (d)  $x = 0.16$ , (e)  $x = 0.18$  and (f)  $x = 0.20$ .



**Fig. S2.** (a)  $d_{33}$  values of PNN- $x$ LN-PT ceramics. (b) Comparison of  $d_{33}$  versus  $T_m$  for various lead-based polynary piezoelectric ceramics with data collected from the reported literature.<sup>1-15</sup>

**Table S1** Rietveld refinements parameters of PNN- $x$ PLN-PT ceramics.

Composition	Structure	Fraction (%)	Lattice parameters	Agreement factors	
				$R_{wp}\%$	$\chi^2$
$x=0.04$	$P4mm$	100	$a=3.9844(\text{\AA})$ $c=4.0279(\text{\AA})$	5.56	2.07
	$R3m$	0	/		
$x=0.08$	$P4mm$	81.8	$a=4.00183(\text{\AA})$ $c=4.0417(\text{\AA})$	4.66	1.49
	$R3m$	18.2	$a=4.0362(\text{\AA})$ $\alpha=89.947^\circ$		
$x=0.12$	$P4mm$	69.5	$a=4.0125(\text{\AA})$ $c=4.0433(\text{\AA})$	4.79	1.56
	$R3m$	30.5	$a=4.0233(\text{\AA})$ $\alpha=89.924^\circ$		
$x=0.16$	$P4mm$	55.1	$a=4.0205(\text{\AA})$ $c=4.0392(\text{\AA})$	4.67	1.51
	$R3m$	44.9	$a=4.0279(\text{\AA})$ $\alpha=89.931^\circ$		
$x=0.18$	$P4mm$	36.5	$a=4.0218(\text{\AA})$ $c=4.0330(\text{\AA})$	4.98	1.69
	$R3m$	63.5	$a=4.0258(\text{\AA})$ $\alpha=89.943^\circ$		
$x=0.20$	$P4mm$	0	/	4.73	1.54
	$R3m$	100	$a=4.0304(\text{\AA})$ $\alpha=89.938^\circ$		

## References

- 1 Y. Zhang, H. Liu, S. Sun, S. Deng and J. Chen, Systematic study of structure and piezoelectric properties of  $\text{Pb}(\text{Ni}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  by in situ synchrotron diffraction, *J. Am. Ceram. Soc.*, 2020, **104**, 604-612.
- 2 D. Shen, X. Li, Z. Wang, Y. Liu, C. He, T. Li, H. Tailor and X. Long, Preparation and characterization of  $(1-x)\text{Pb}(\text{Lu}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-}x\text{PbTiO}_3$  binary ferroelectric ceramics with high Curie temperature, *Mater. Lett.*, 2012, **84**, 1-4.
- 3 E. F. Alberta and A. S. Bhalla, High strain and low mechanical quality factor piezoelectric  $\text{Pb}[(\text{Sc}_{1/2}\text{Nb}_{1/2})_{0.575}\text{Ti}_{0.425}]\text{O}_3$  ceramics. *Mater. Lett.*, 1998, **35**, 199-201.
- 4 E. F. Alberta and A. S. Bhalla, Piezoelectric Properties of  $\text{Pb}(\text{InNb})_{1/2}\text{O}_3$  -  $\text{PbTiO}_3$  Solid Solution Ceramics, *J. Korean Phys. Soc.*, 1998, **32**, S1265-S1267.
- 5 H. Tang, M. F. Zhang, S. J. Zhang, Y. J. Feng, F. Li and T. R. ShROUT, Investigation of dielectric and piezoelectric properties in  $\text{Pb}(\text{Ni}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbHfO}_3\text{-PbTiO}_3$  ternary system, *J. Eur. Ceram. Soc.*, 2013, **33**, 2491-2497.
- 6 Y. Chen, X. Zhang, J. Pan and K. Chen, Study of the structure and electrical properties of PMN-PNN-PT ceramics near the morphotropic phase boundary, *J. Electroceramics*, 2006, **16**, 109-114.
- 7 H. Tang, S. Zhang, Y. Feng, F. Li, T. R. ShROUT and D. Johnson, Piezoelectric Property and Strain Behavior of  $\text{Pb}(\text{Yb}_{0.5}\text{Nb}_{0.5})\text{O}_3\text{-PbHfO}_3\text{-PbTiO}_3$  Polycrystalline Ceramics, *J. Am. Ceram. Soc.*, 2013, **96**, 2857-2863.
- 8 D. Wang, M. Cao, S. Zhang and J. L. Jones, Investigation of Ternary System  $\text{PbHfO}_3\text{-PbTiO}_3\text{-Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$  with Morphotropic Phase Boundary Compositions, *J. Am. Ceram. Soc.*, 2012, **95**, 3220-3228.
- 9 G. Zhu, H. Liu, S. Sun, B. Gao and J. Chen, Characterization and high piezoelectric performance of  $\text{Pb}(\text{Fe}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-Pb}(\text{In}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-PbTiO}_3$  ternary ceramics, *Inorg. Chem. Front.*, 2019, **6**, 3070-3076.
- 10 Y. Zhang, H. Liu, S. Sun, Y. Liu, C. Huo, H. Qi, S. Deng and J. Chen, High Piezoelectric Performance in  $\text{Pb}(\text{Ni}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-Pb}(\text{Sc}_{1/2}\text{Nb}_{1/2})\text{O}_3\text{-PbTiO}_3$

- Ternary System Featuring Small Structural Distortion and Heterogeneous Domain Configuration, *ACS Appl. Mater. Interfaces*, 2022, **14**, 13528-13538.
- 11 X. Luo, J. Zeng, X. Shi, L. Zheng, K. Zhao, Z. Man and G. Li, Dielectric, ferroelectric and piezoelectric properties of MnO<sub>2</sub>-doped Pb(Yb<sub>1/2</sub>Nb<sub>1/2</sub>)O<sub>3</sub>-Pb(Zr,Ti)O<sub>3</sub> ceramics, *Ceram. Int.*, 2018, **44**, 8456-8460.
- 12 D. Lin, S. Zhang, E. Gorzkowski, S. Zhou, W. Liu and F. Li, Investigation of morphotropic phase boundaries in PIN-PSN-PT relaxor ferroelectric ternary systems with high  $T_{r-t}$  and  $T_c$  phase transition temperatures, *J. Eur. Ceram. Soc.*, 2017, **37**, 2813-2823.
- 13 D. Wang, M. Cao and S. Zhang, Phase diagram and properties of Pb(In<sub>1/2</sub>Nb<sub>1/2</sub>)O<sub>3</sub>-Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-PbTiO<sub>3</sub> polycrystalline ceramics, *J. Eur. Ceram. Soc.*, 2012, **32**, 433-439.
- 14 T. Li, X. Li, D. Guo, Z. Wang, Y. Liu, C. He, T. Chu, L. Ai, D. Pang, X. Long and T. A. Vanderah, Phase Diagram and Properties of High  $T_C/T_{R-T}$  Pb(In<sub>1/2</sub>Nb<sub>1/2</sub>)O<sub>3</sub>-Pb(Zn<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-PbTiO<sub>3</sub> Ferroelectric Ceramics, *J. Am. Ceram. Soc.*, 2013, **96**, 1546-1553.
- 15 K. Wen, J. Qiu, H. Ji, K. Zhu, J. Liu, J. Wang, J. Du and F. Zhu, Investigation of phase diagram and electrical properties of  $x$ Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-(1- $x$ )Pb(Zr<sub>0.4</sub>Ti<sub>0.6</sub>)O<sub>3</sub> ceramics, *J. Mater. Sci. Mater. Electron.*, 2014, **25**, 3003-3009.