



中國科學院強磁場科學中心

High Magnetic Field Laboratory, Chinese Academy of Sciences



Fabrication and Characterization of Epitaxial KNN Films

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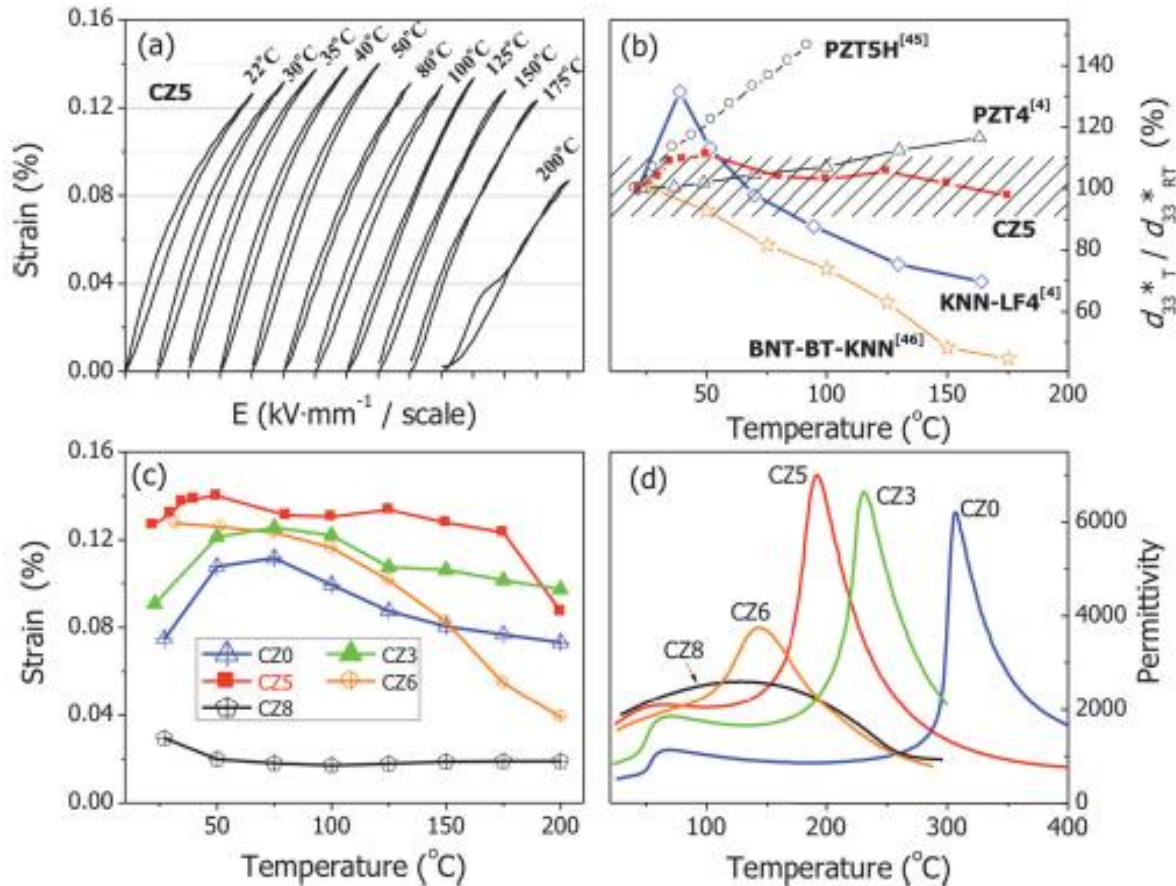
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Shunyi Li and Andreas Klein

Department of Material Science, Technical University of Darmstadt, Darmstadt, Germany

Temperature-Insensitive (K,Na)NbO₃-Based Lead-Free Piezoactuator Ceramics

Ke Wang,* Fang-Zhou Yao, Wook Jo, Danka Gobeljic, Vladimir V. Shvartsman, Doru C. Lupascu, Jing-Feng Li, and Jürgen Rödel



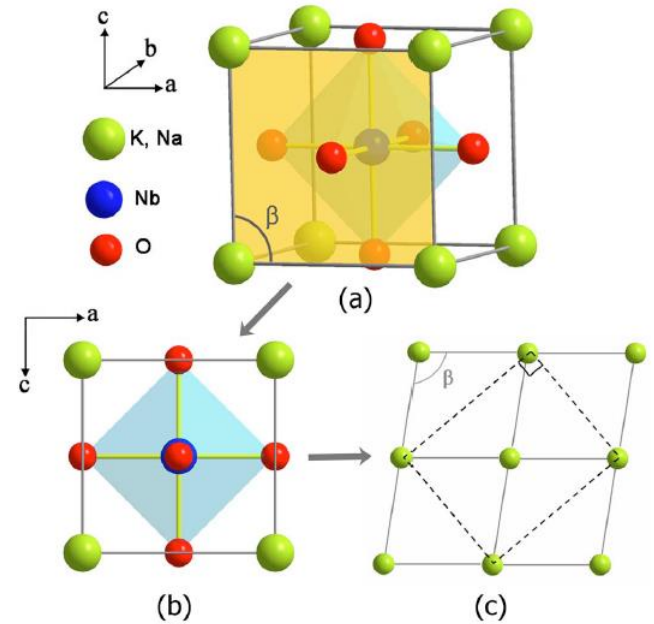
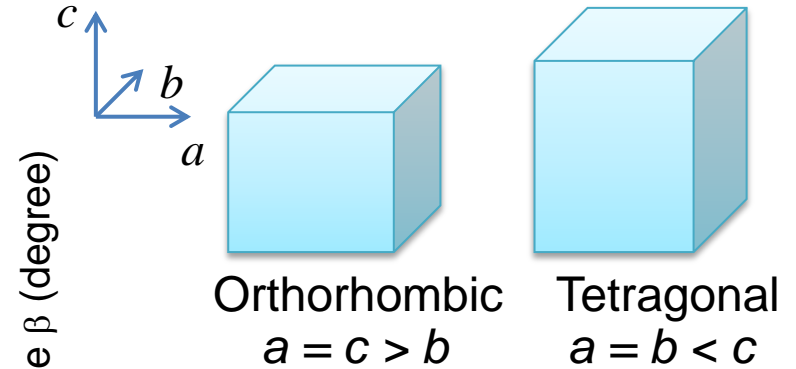
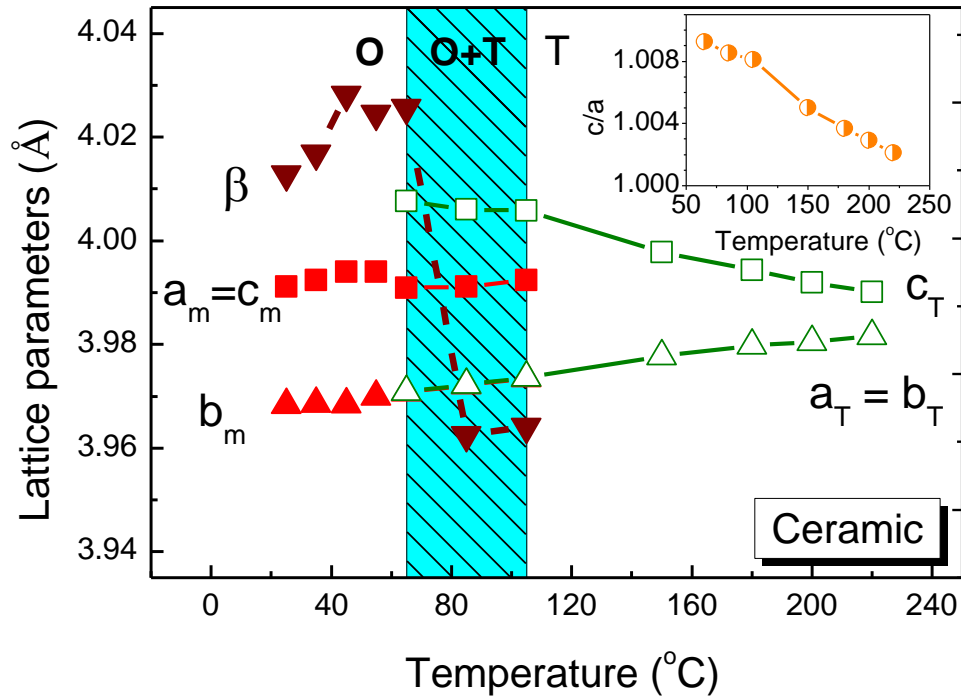
KNN: 0.95(Na_{0.49}K_{0.49}Li_{0.02})(Nb_{0.8}Ta_{0.2})O₃-0.05CaZrO₃ with 2 wt% MnO₂

Outline



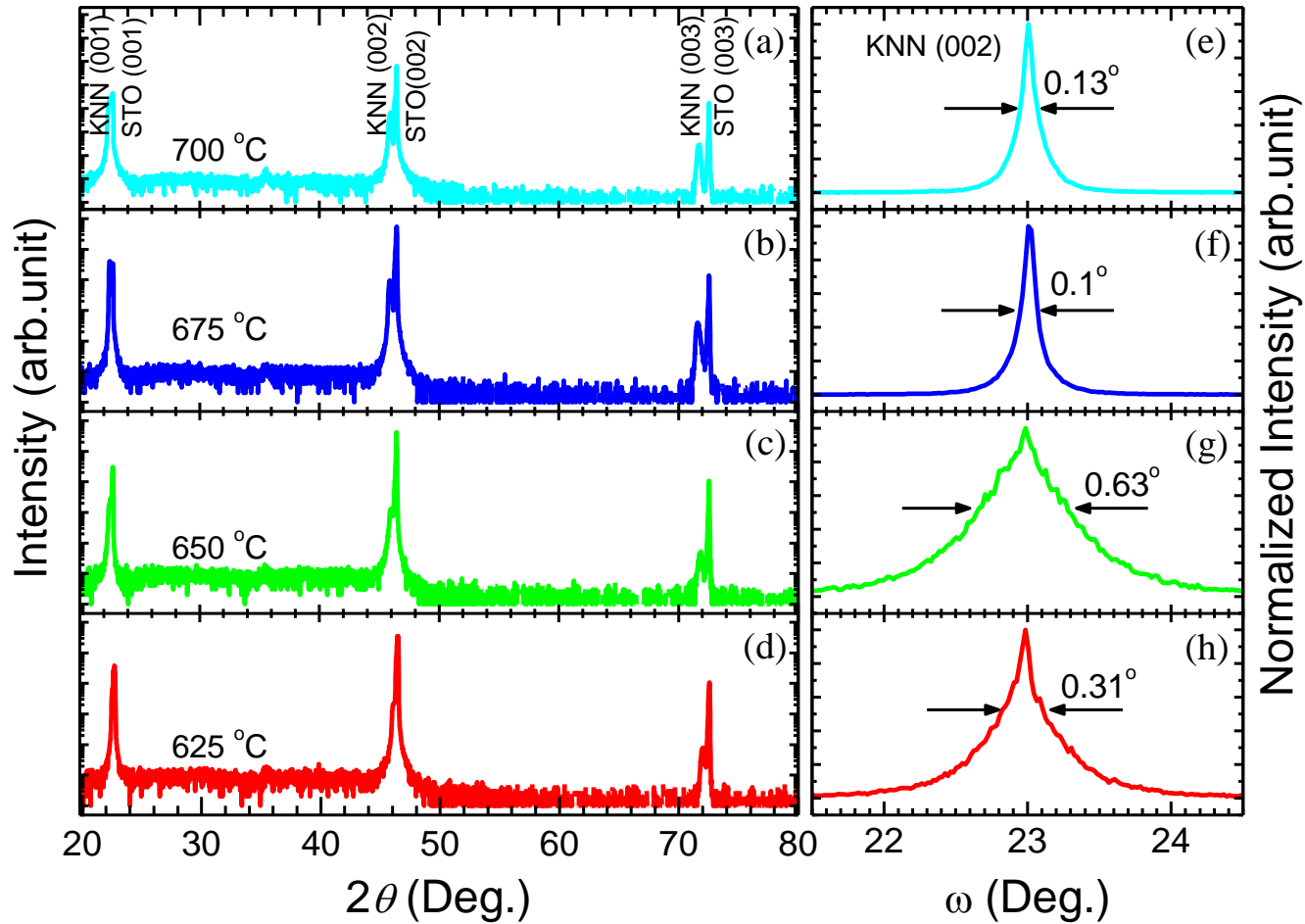
- Epitaxial KNN films growth with pulsed laser deposition (PLD)
- High fatigue resistance of KNN films
- KNN/Pt and PZT/Pt interfaces studied with X-ray photoelectron spectroscopy (XPS)

Lattice constant of KNN ceramic



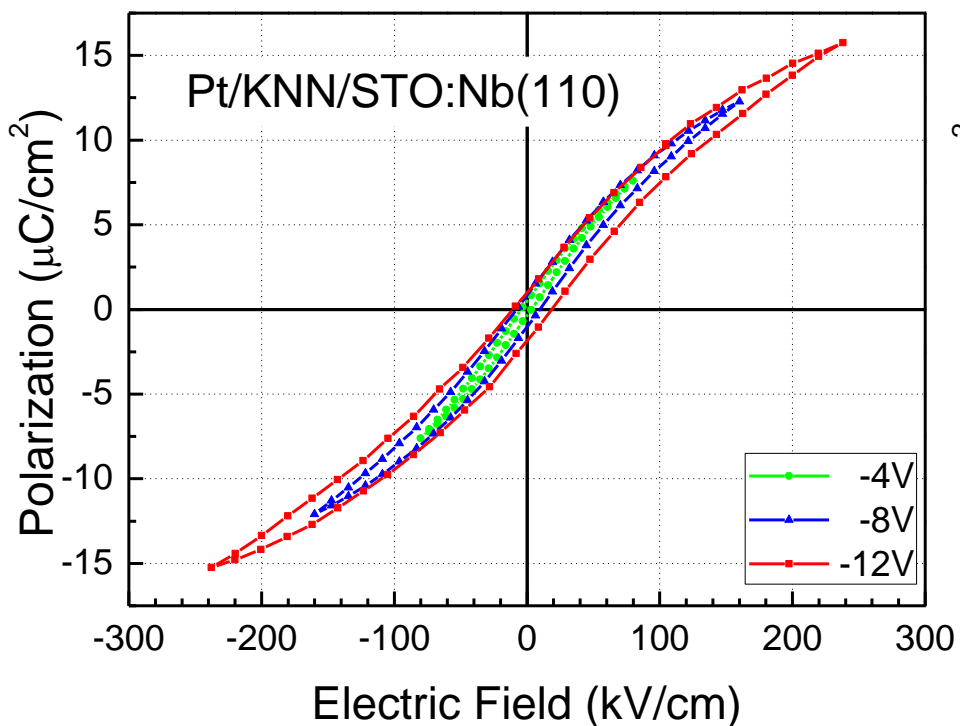
LSMO: $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ $a_{\text{LSMO}} = 3.873 \text{ \AA}$
 LSSO: $\text{La}_{0.07}\text{Sr}_{0.93}\text{SnO}_3$ $a_{\text{LSSO}} = 4.035 \text{ \AA}$
 STO: SrTiO_3 $a_{\text{STO}} = 3.905 \text{ \AA}$

KNN film on STO:Nb (001) single crystalline

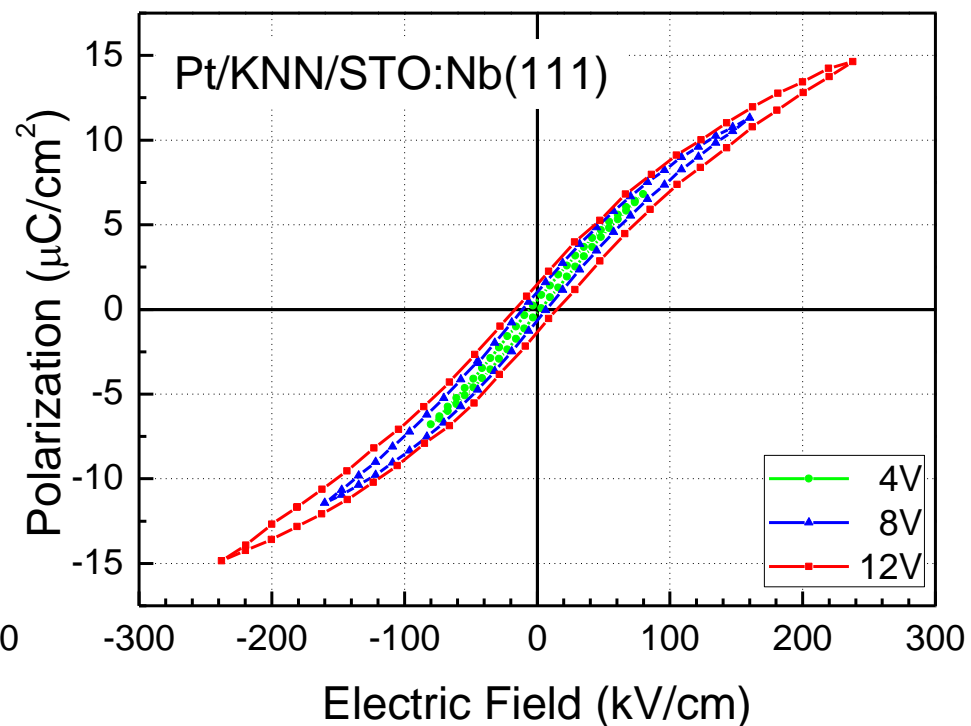


High quality epitaxial KNN films can be obtained on STO:Nb substrate

Ferroelectric properties of KNN/NSTO films

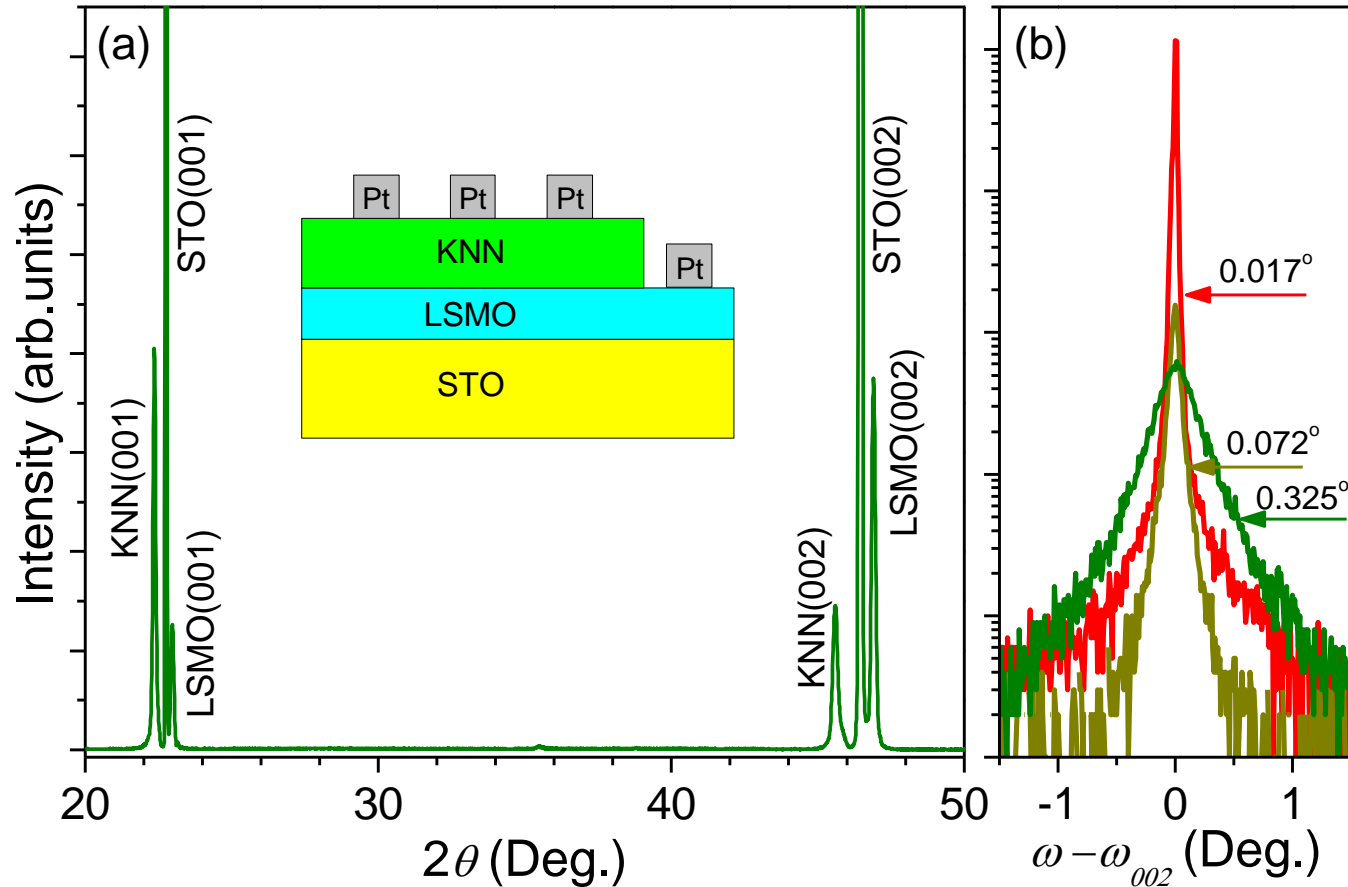


Slim hysteresis



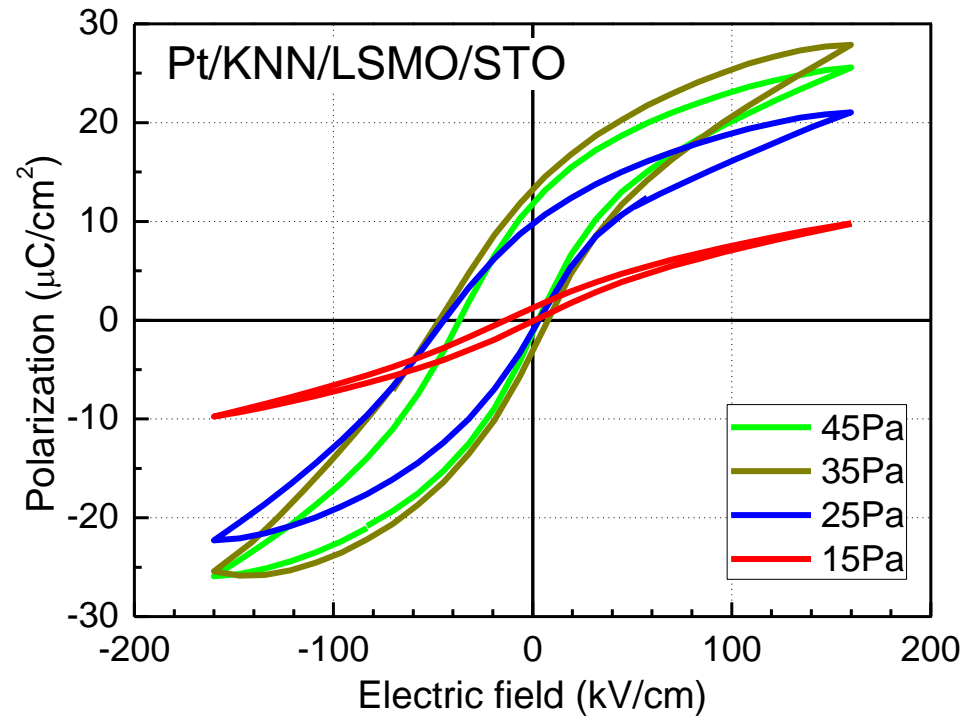
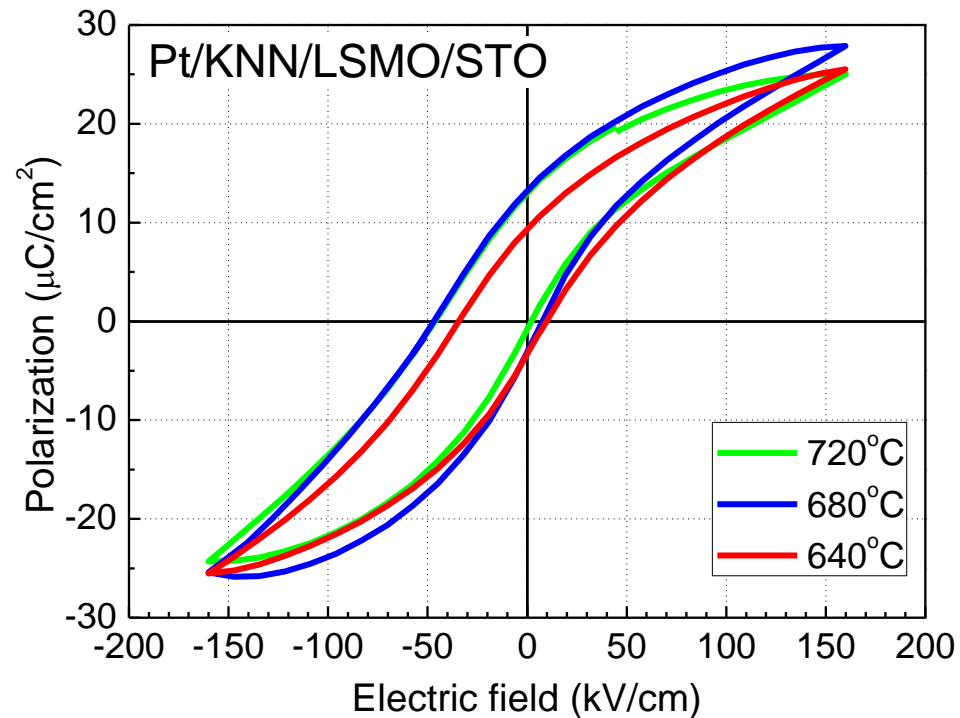
Small E_c and P_r

Epitaxial KNN films grown on conductive oxide electrode



- Epitaxial film since only 00l reflections were recorded
- A broad rocking curve of KNN (002) peak

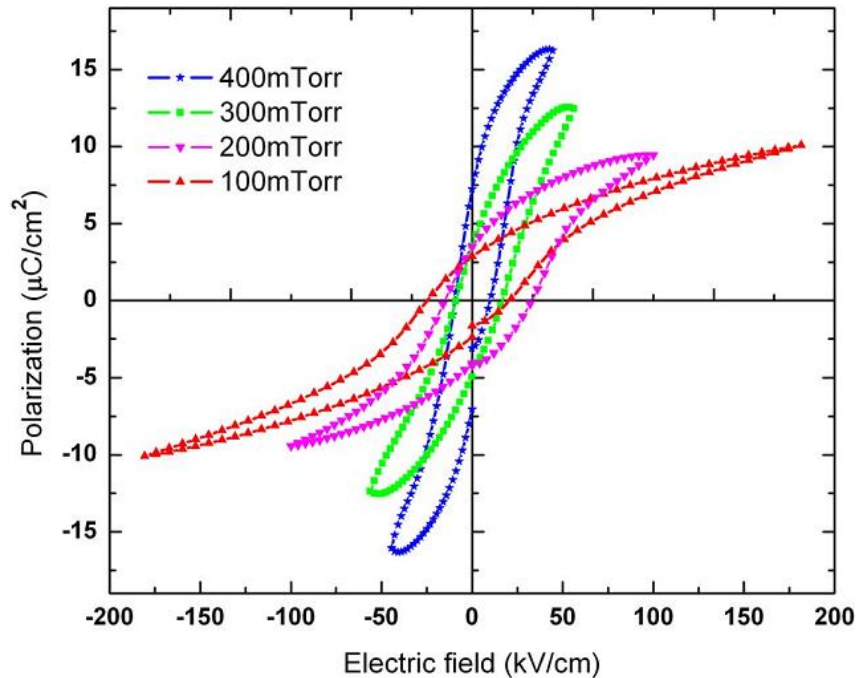
Ferroelectric properties of KNN films



- The saturated P-E loops are measured at 1k Hz
- The asymmetric loop might be induced by the asymmetric interfaces

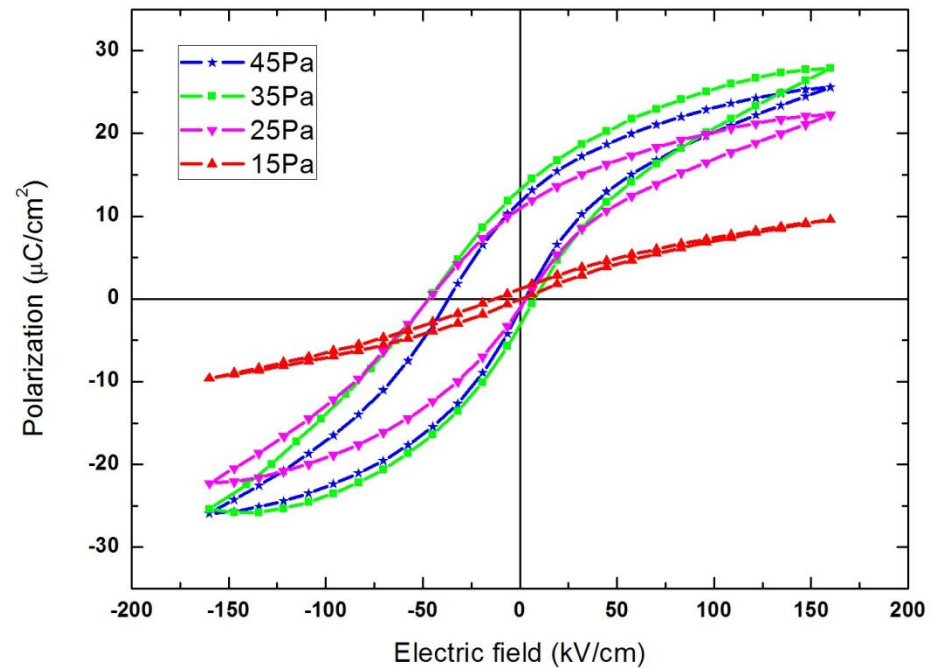
Hysteresis of KNN film capacitor

SrRuO₃/KNN/SrRuO₃/STO(001)



M. Abazari *et al.*, APL 93, 192910 (2008).

Pt/KNN/LSMO/STO(001)

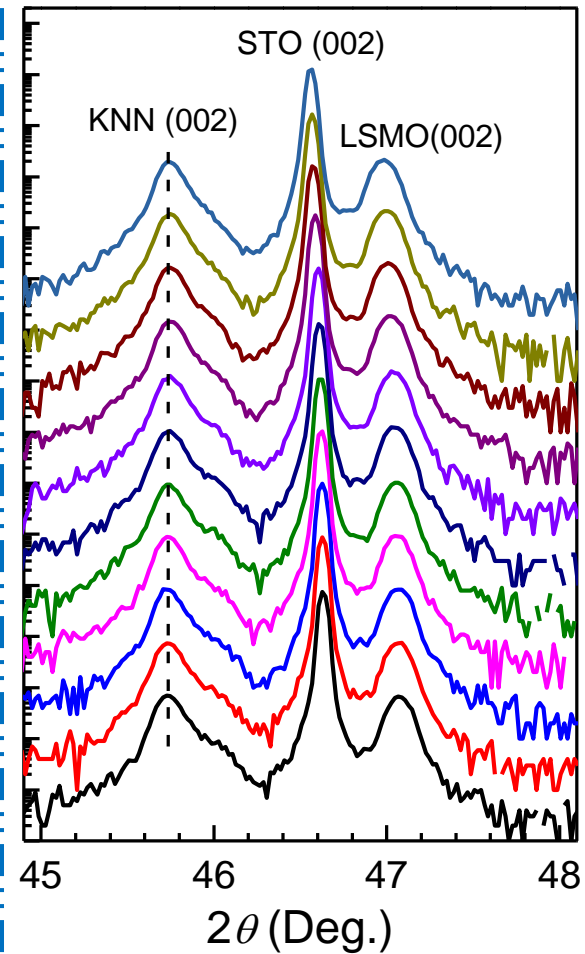
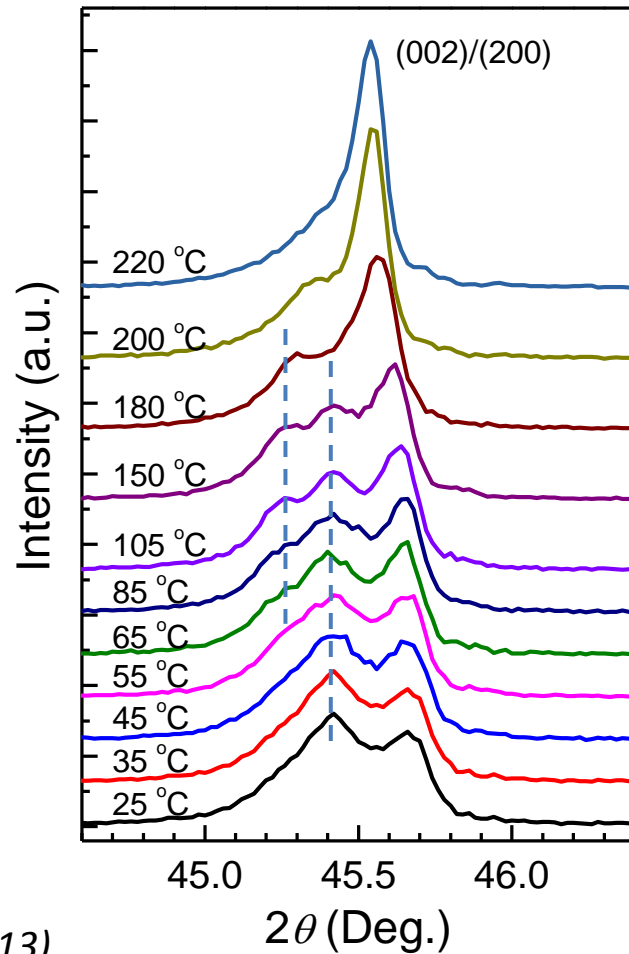
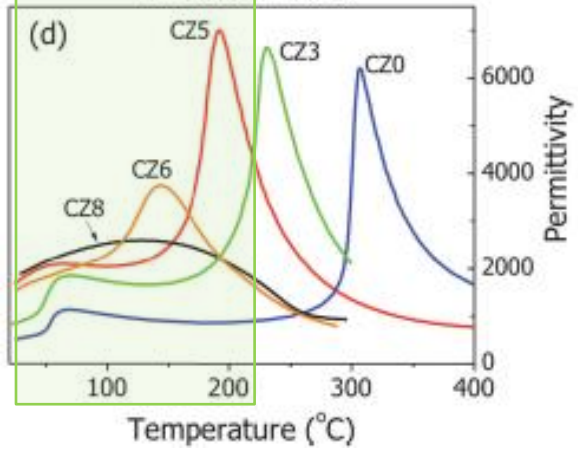
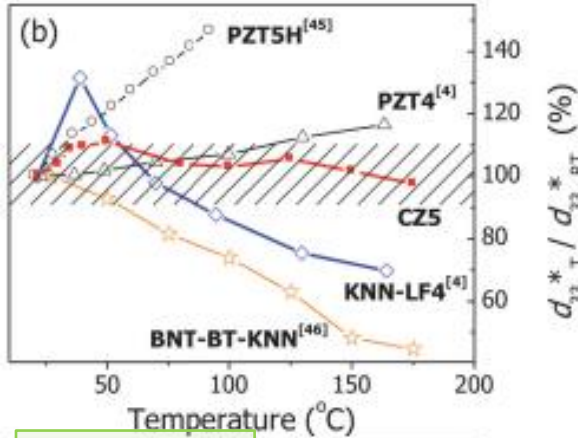


- $2Pr \sim 15 \mu\text{C}/\text{cm}^2$
- Higher P_s and saturated loops

Structure variation of KNN films with temperature

Ceramic

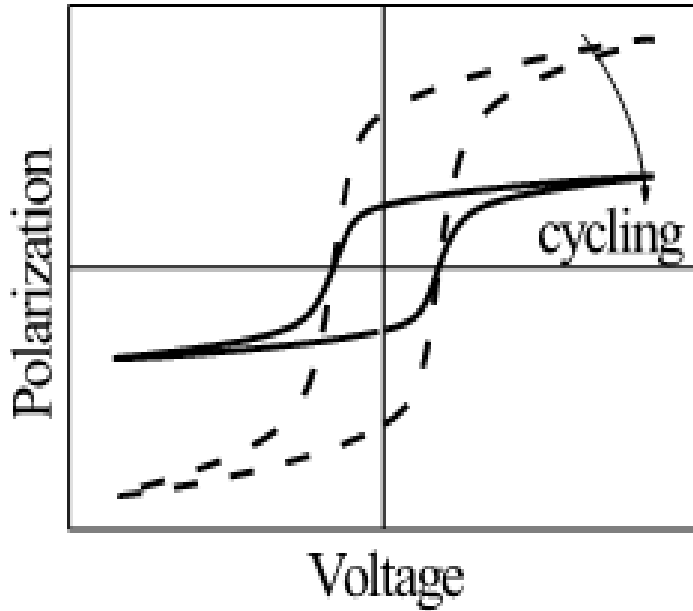
Film



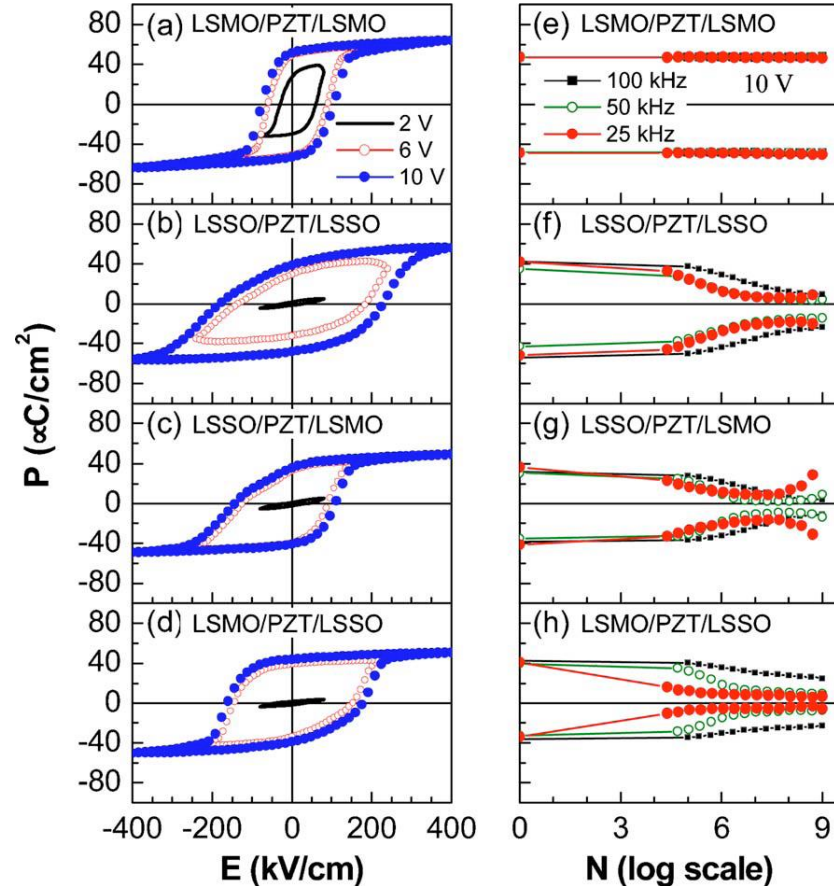
Adv. Funct. Mater., 23, 4079-86 (2013).

The KNN films might have a even higher thermal stability than that of the ceramic

Fatigue of PZT films



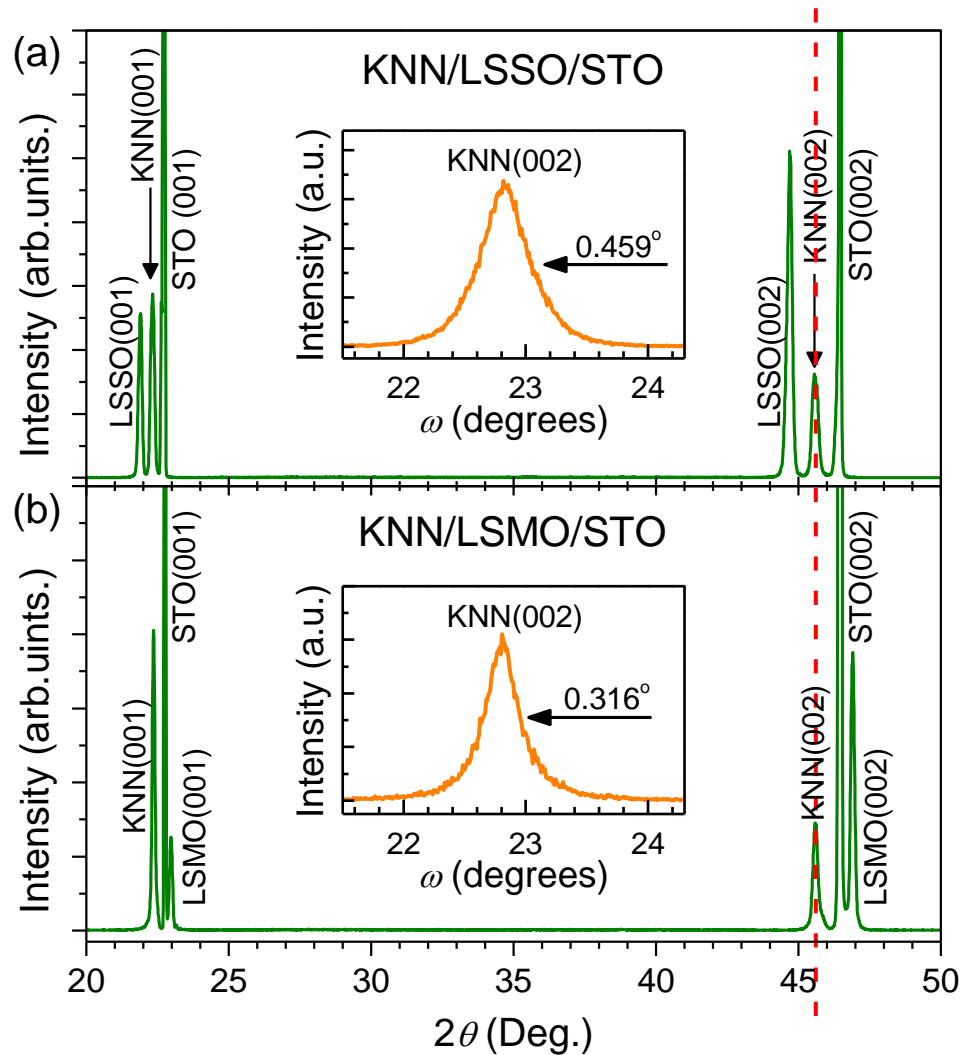
Fatigue



F. Chen et al. APL, 90, 192907 (2007)

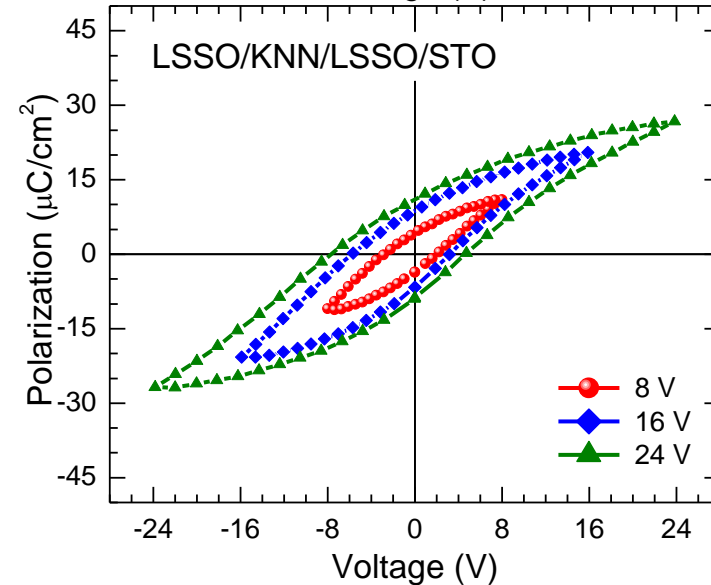
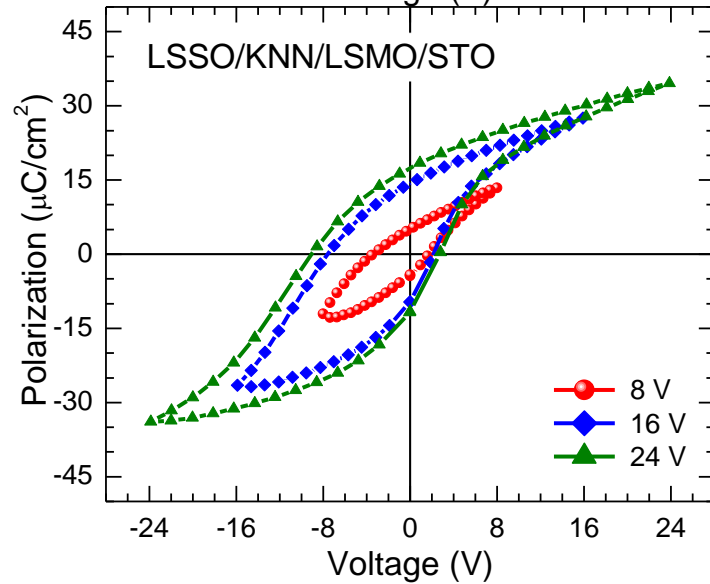
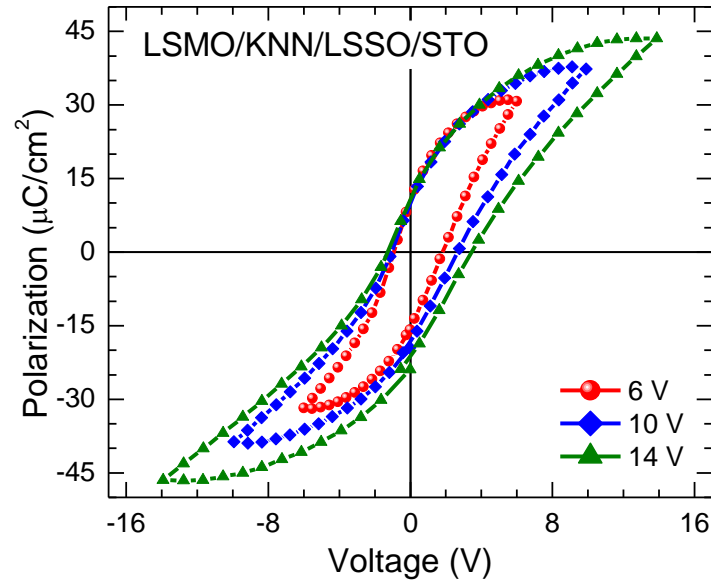
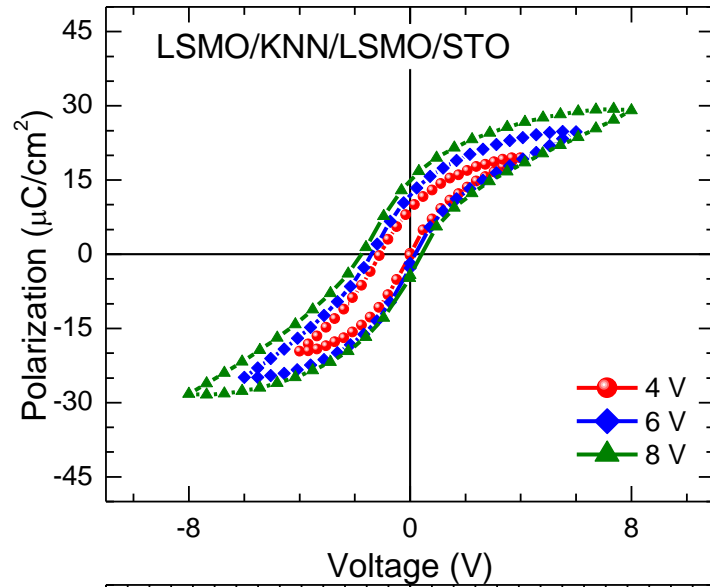
The fatigue behavior of PZT epitaxial films highly depends on the (p/n)-type of the conductive oxide electrode

Epitaxial KNN films grown on conductive oxide electrode

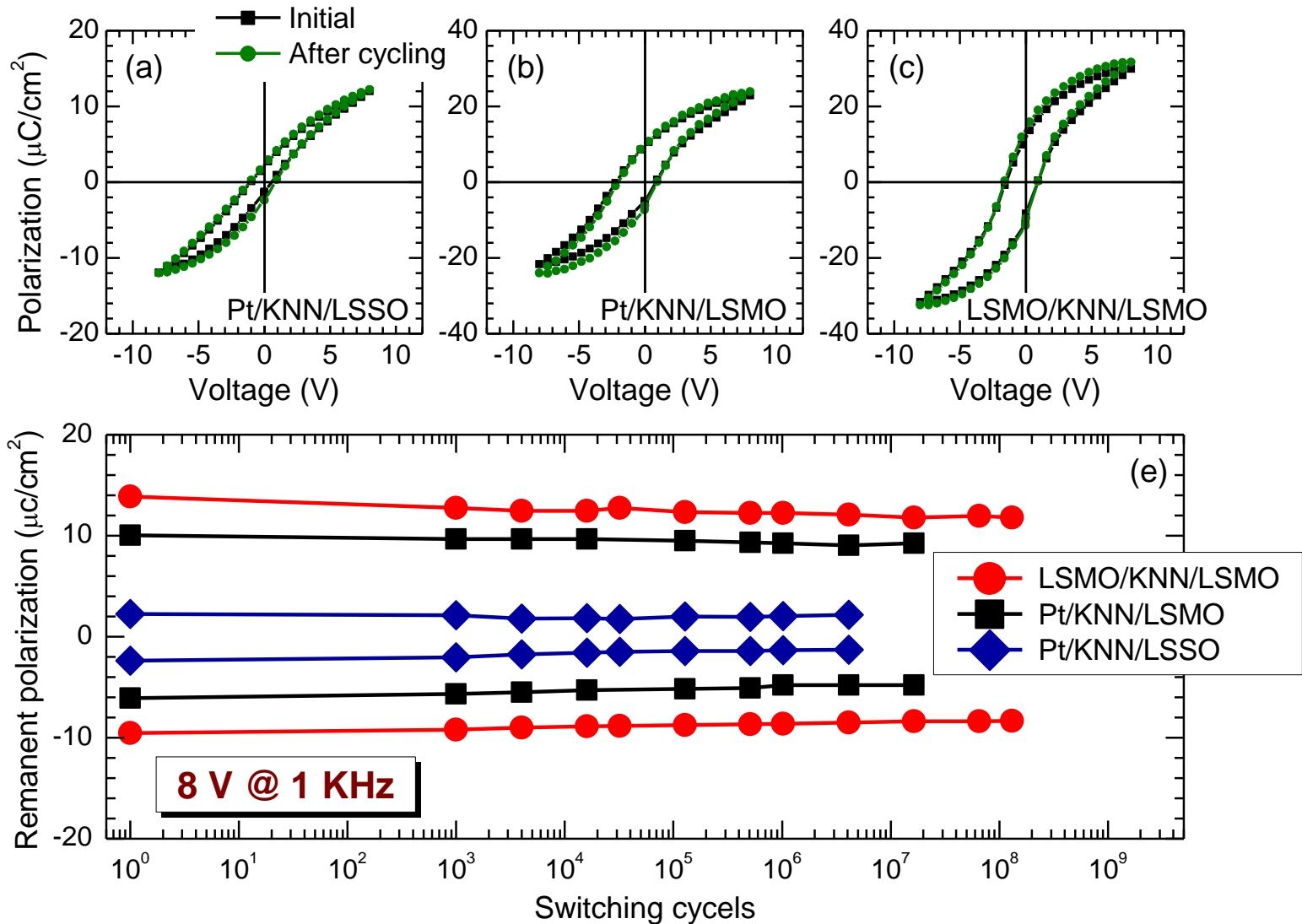


The strain in the films is relaxed reflecting by the same KNN (002) position

P-E loops of KNN films

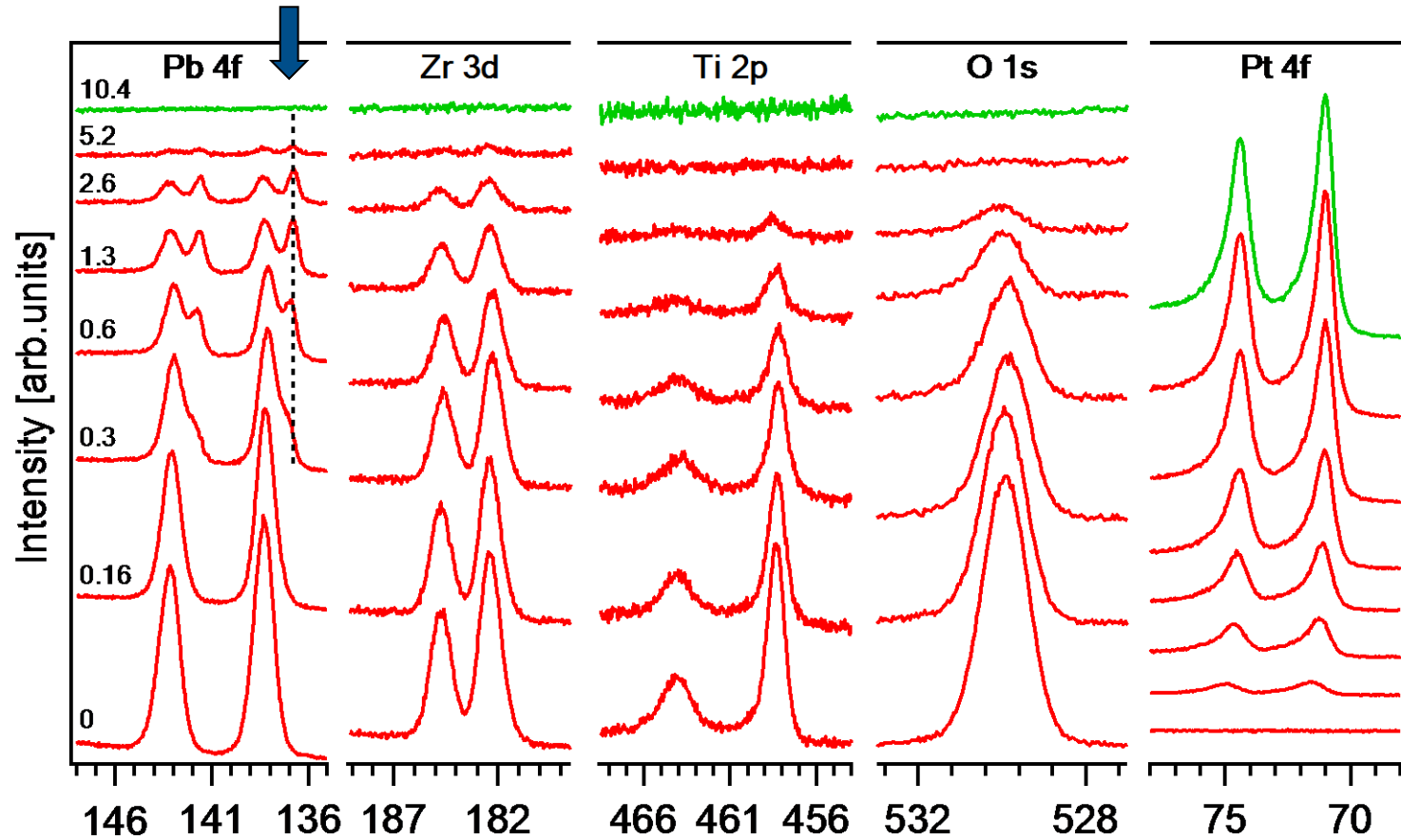


Fatigue of KNN films



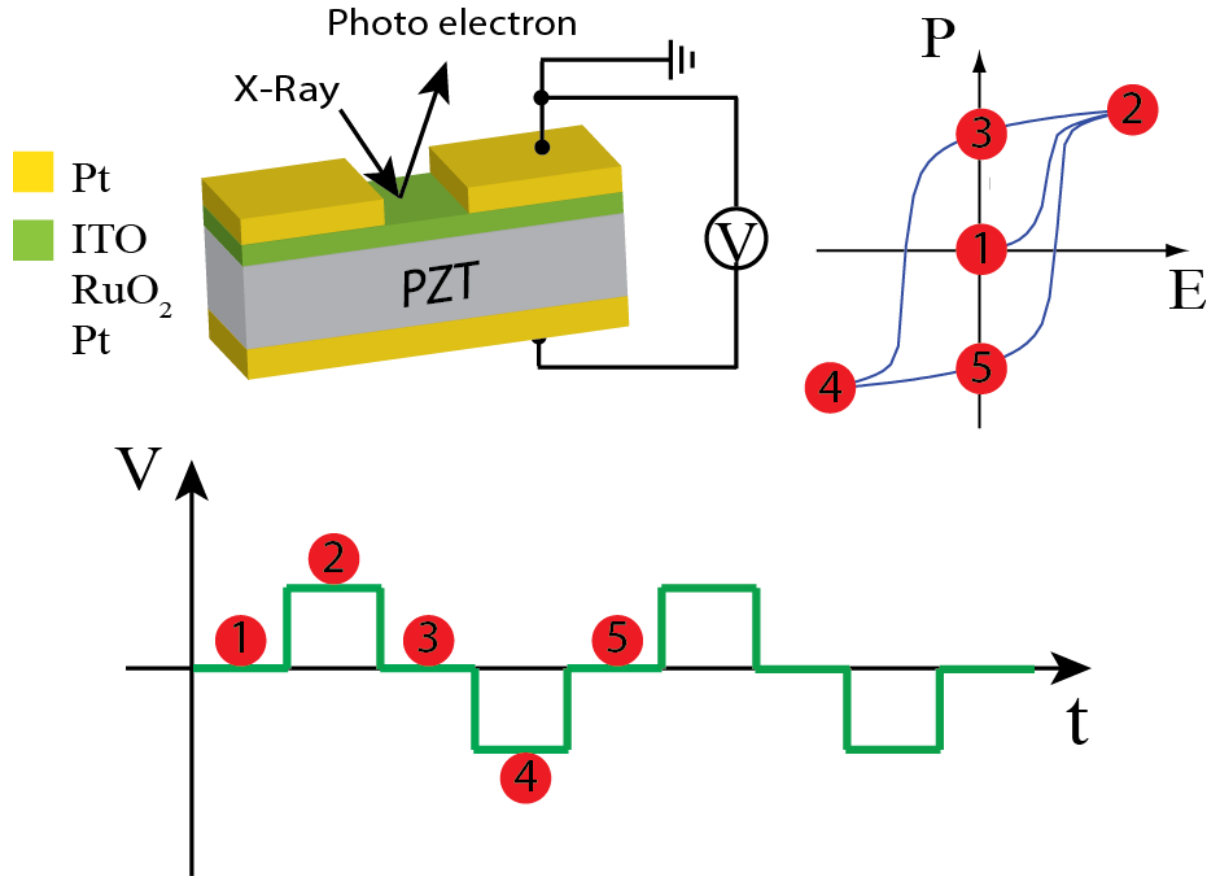
KNN films show a high fatigue resistance under bipolar cycling

Reaction at PZT/Pt interface

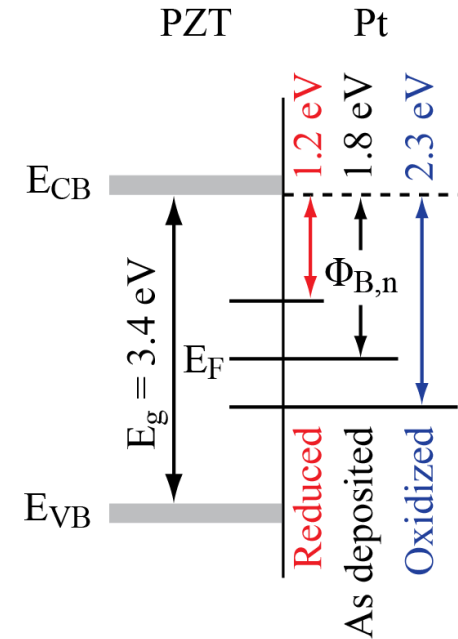
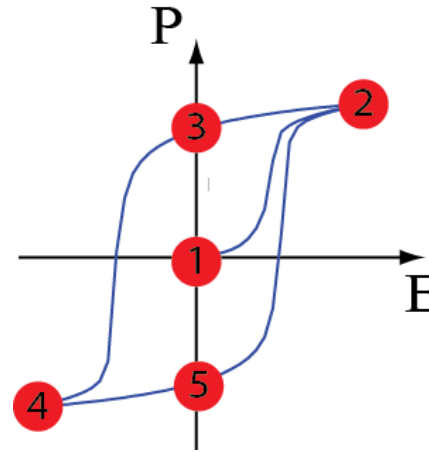
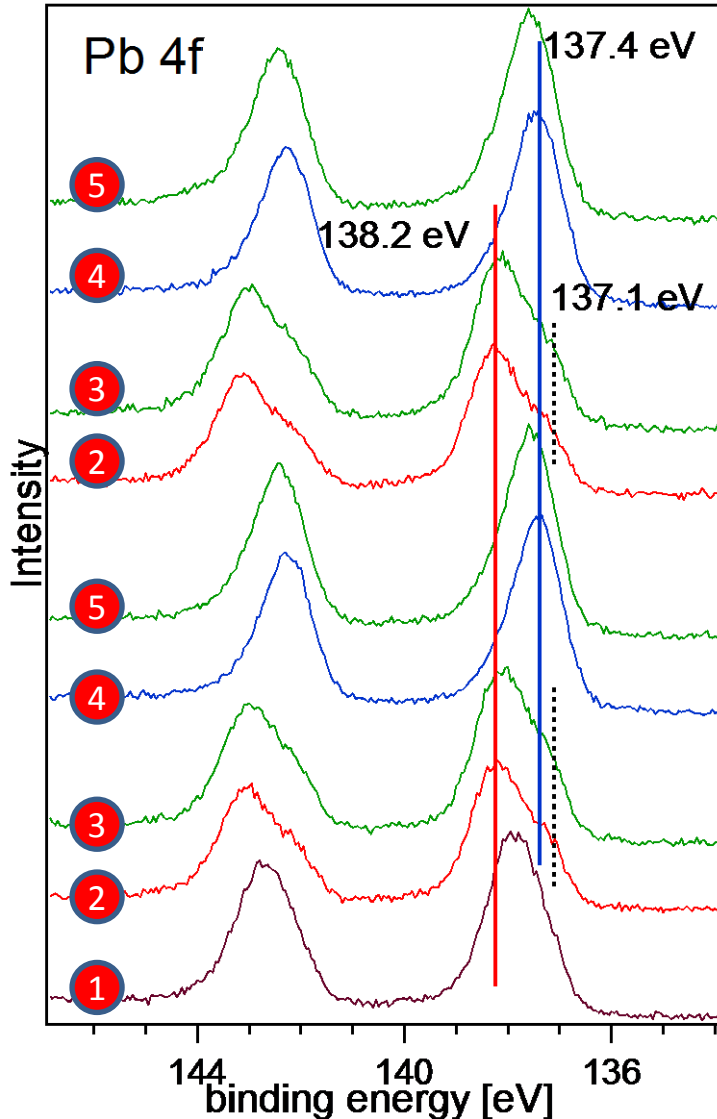


Chemical decomposition of PZT during Pt deposition

In-situ XPS measurement in poling process

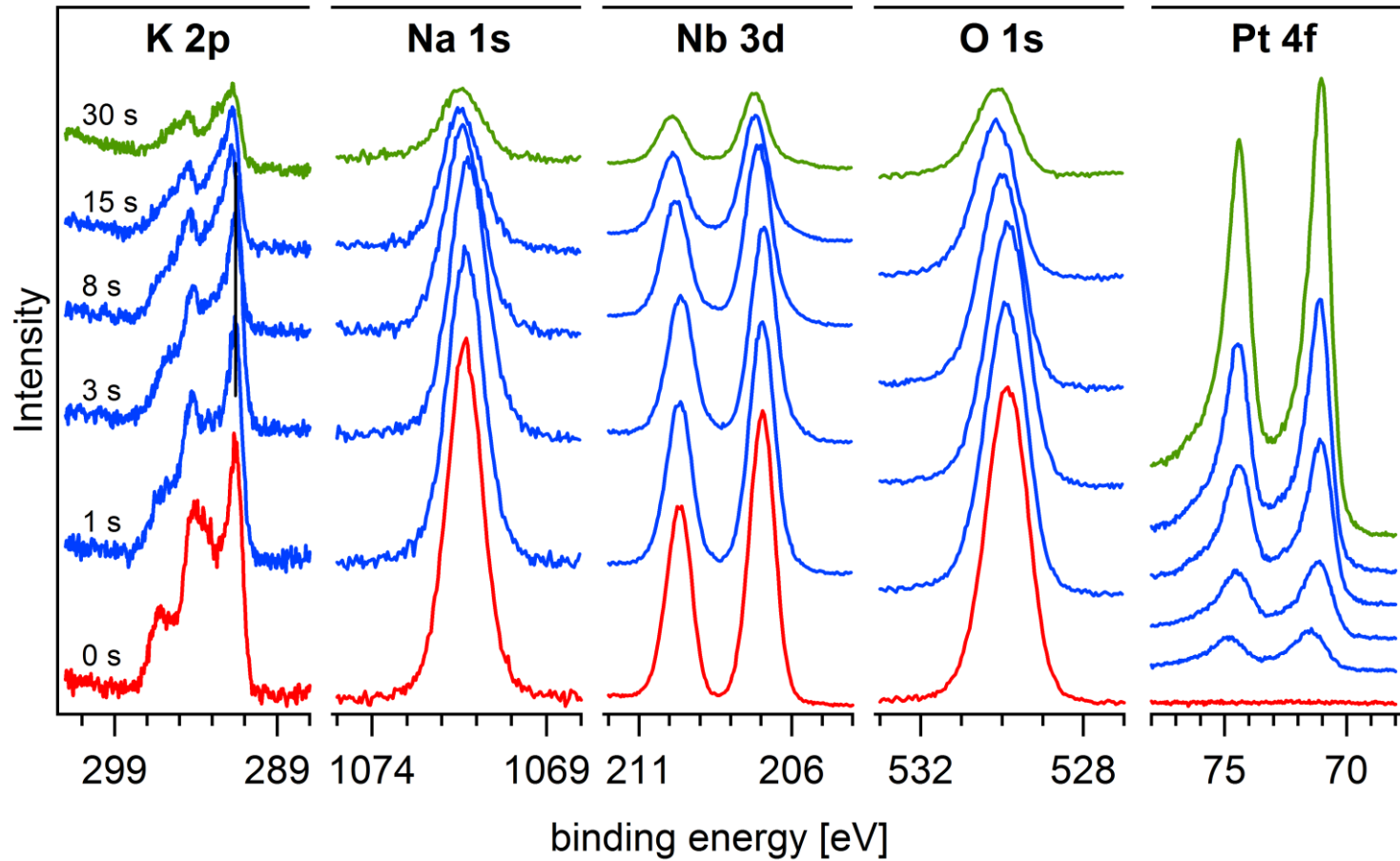


E_F variation at PZT/Pt interface during switching



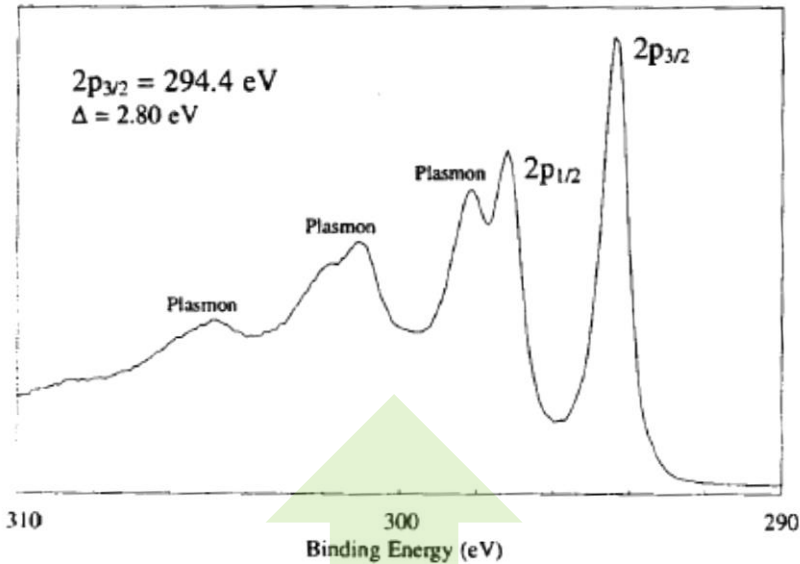
- PZT/Pt interface is weakly *p*-type
- Positive state, E_F shift upward, acceptor defects appear

KNN/Pt interface

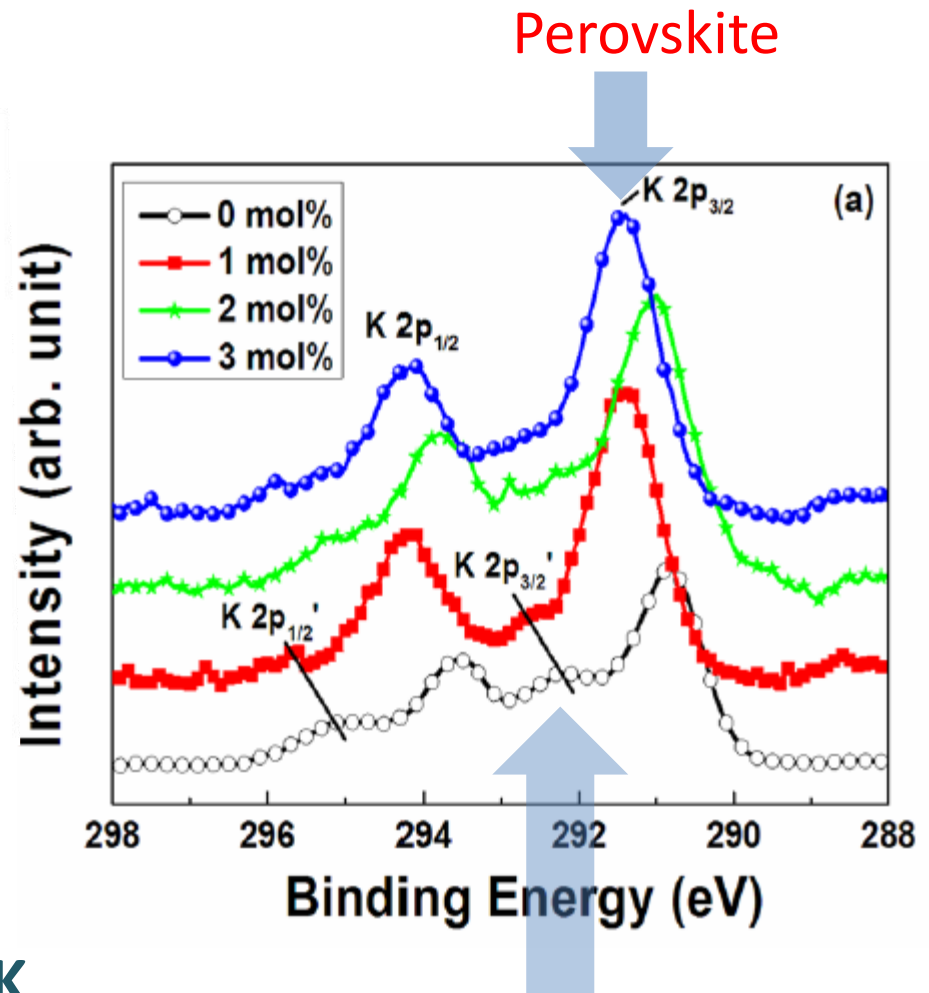


Obvious decomposition of KNN during Pt deposition was **Not** found

KNN/Pt interface

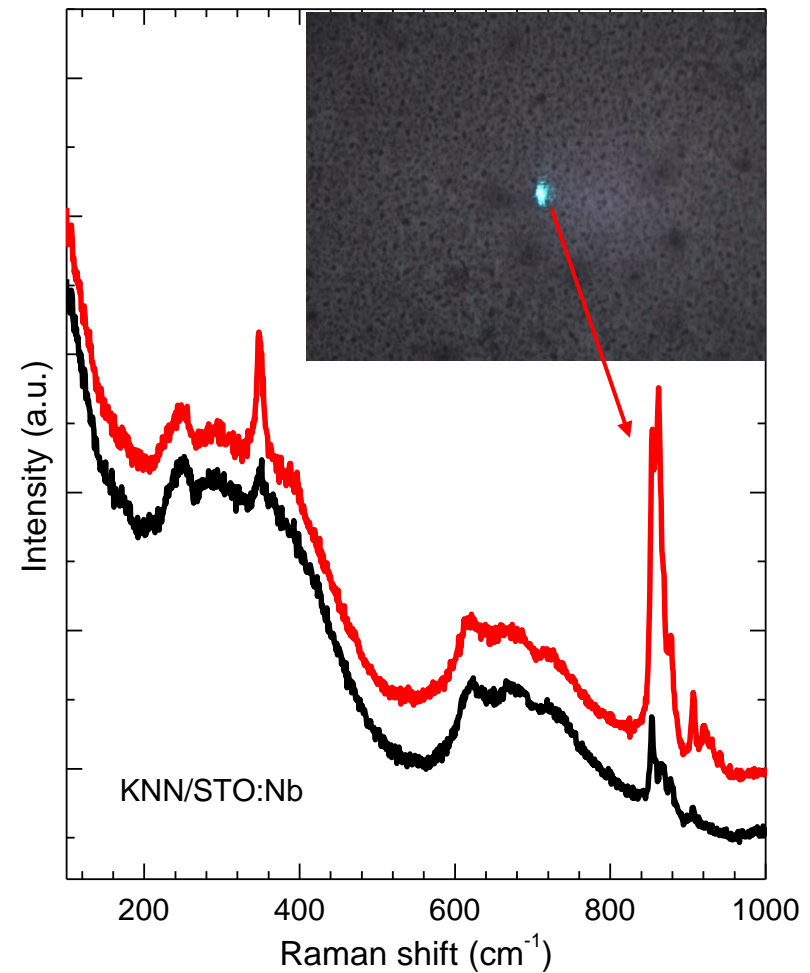
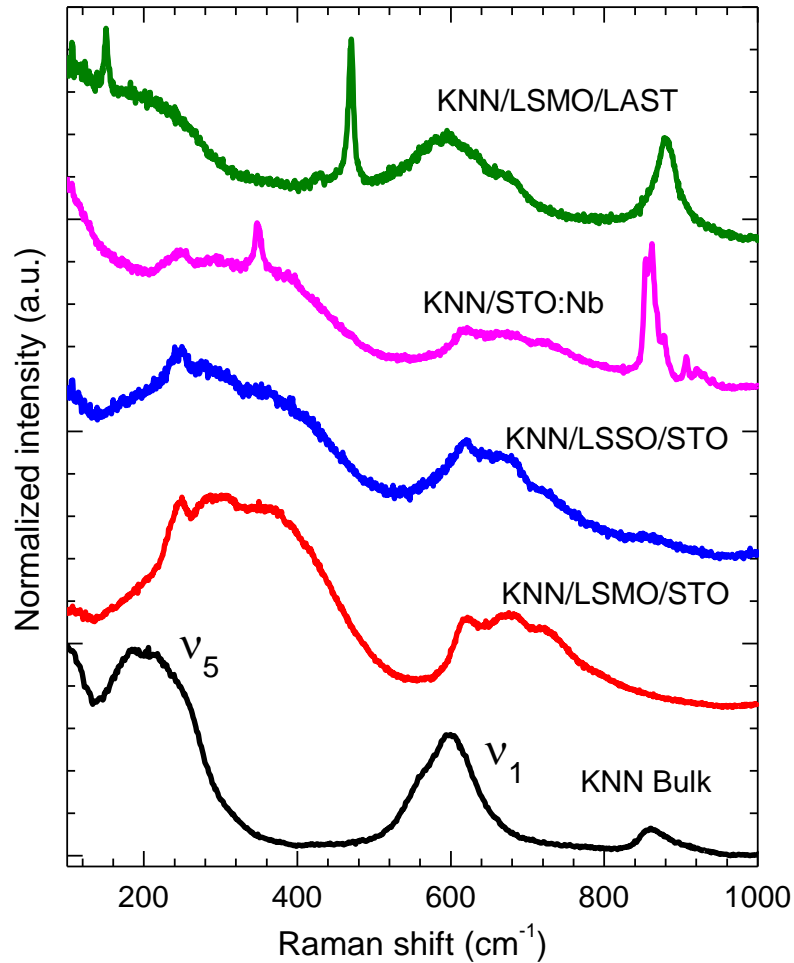


Plasmon peak with metallic K



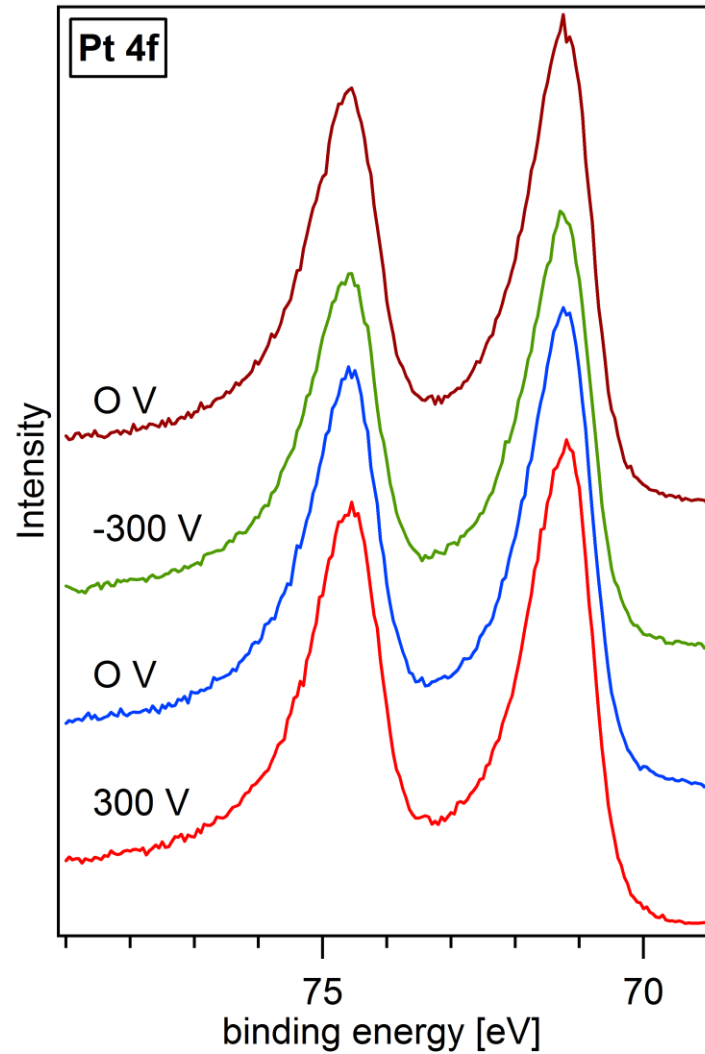
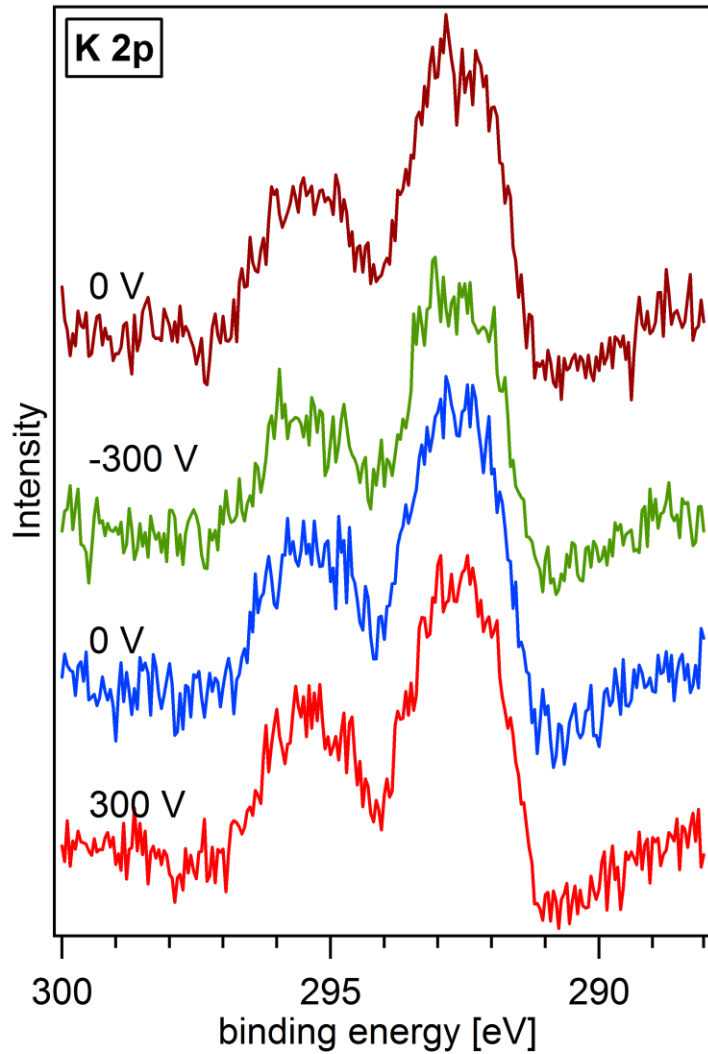
Non-Perovskite

Raman scattering spectrum



Raman shift of the KNN epitaxial films is sensitive to the electrode and substrate used

KNN/Pt interface (switching)



No binding energy shift and KNN decomposition during switching

Conclusions

- **KNN epitaxial films can be fabricated on conductive oxide substrates with PLD**
- **Their crystalline quality and ferroelectric property depend on the substrate (electrode)**
- **Epitaxial KNN films might have a even higher thermal stability**
- **KNN films show a high fatigue resistance behavior during bipolar switching, which might be induced by the phase separation at the surface**

Thanks for you attention!