Fabrication and Characterization of Epitaxial KNN Films

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

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KNN: $0.95(\text{Na}_{0.49}\text{K}_{0.49}\text{Li}_{0.02})(\text{Nb}_{0.8}\text{Ta}_{0.2})\text{O}_3 - 0.05\text{CaZrO}_3$ 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

**Lattice parameters**

<table>
<thead>
<tr>
<th>Material</th>
<th>Formula</th>
<th>Lattice Constant (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LSMO</td>
<td>La$<em>{0.7}$Sr$</em>{0.3}$MnO$_3$</td>
<td>$a_{\text{LSMO}} = 3.873$ Å</td>
</tr>
<tr>
<td>LSSO</td>
<td>La$<em>{0.07}$Sr$</em>{0.93}$SnO$_3$</td>
<td>$a_{\text{LSSO}} = 4.035$ Å</td>
</tr>
<tr>
<td>STO</td>
<td>SrTiO$_3$</td>
<td>$a_{\text{STO}} = 3.905$ Å</td>
</tr>
</tbody>
</table>

**Ceramic Phase Diagram**

- **Orthorhombic**: $a = c > b$
- **Tetragonal**: $a = b < c$

**Angle $\beta$ (degree)**

High quality epitaxial KNN films can be obtained on STO:Nb substrate.
Ferroelectric properties of KNN/NSTO films

Pt/KNN/STO:Nb(110)

Pt/KNN/STO:Nb(111)

Slim hysteresis

Small \( E_c \) and \( Pr \)
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

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

- $2Pr \sim 15 \, \mu\text{C/cm}^2$
- Higher $P_s$ and saturated loops
Structure variation of KNN films with temperature

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

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

F. Chen et al. APL, 90, 192907 (2007)
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
Obvious decomposition of KNN during Pt deposition was Not found
KNN/Pt interface

Plasmon peak with metallic K

Non-Perovskite

Perovskite

Wang et al. JAP, 115, 034104 (2014)
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 even higher thermal stability
- KNN films show high fatigue resistance behavior during bipolar switching, which might be induced by phase separation at the surface

Thanks for your attention!