Winter 3-6-2016

Influence of an electric field on grain growth and sintering in strontium titanate

Wolfgang Rheinheimer
Karlsruhe Institute of Technology, wolfgang.rheinheimer@kit.edu

Fabian Lemke
, Karlsruhe Institute of Technology

Michael Hoffman
, Karlsruhe Institute of Technology

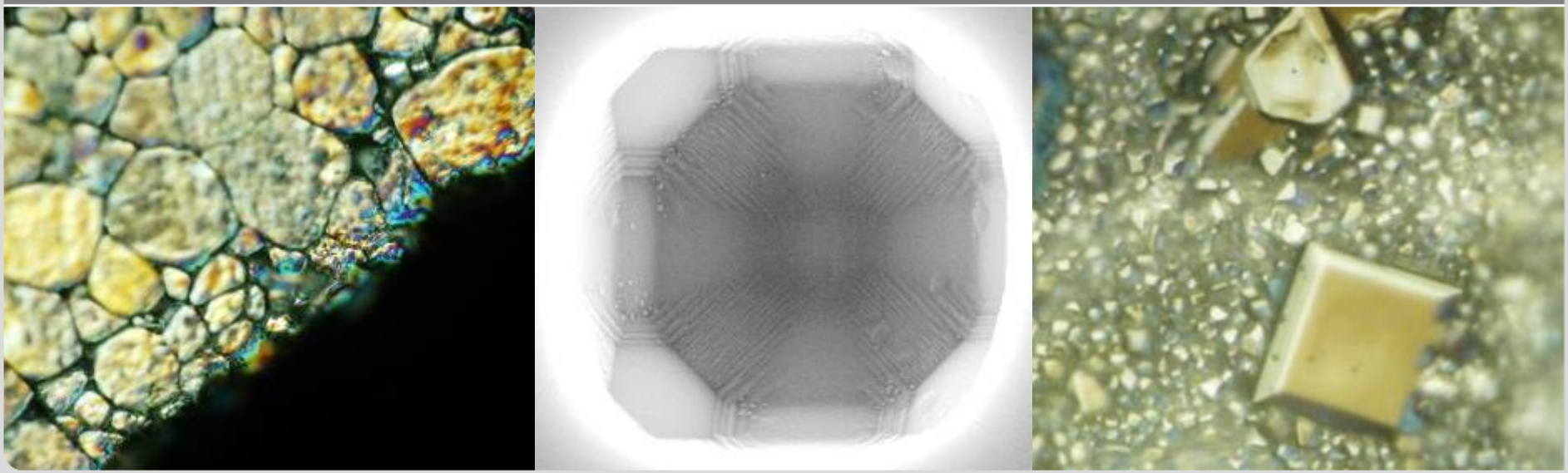
Karlsruhe Institute of Technology

Institute for Applied Materials

Impact of electric fields on grain growth in $SrTiO_3$

Fabian Lemke, Wolfgang Rheinheimer and Michael J. Hoffmann

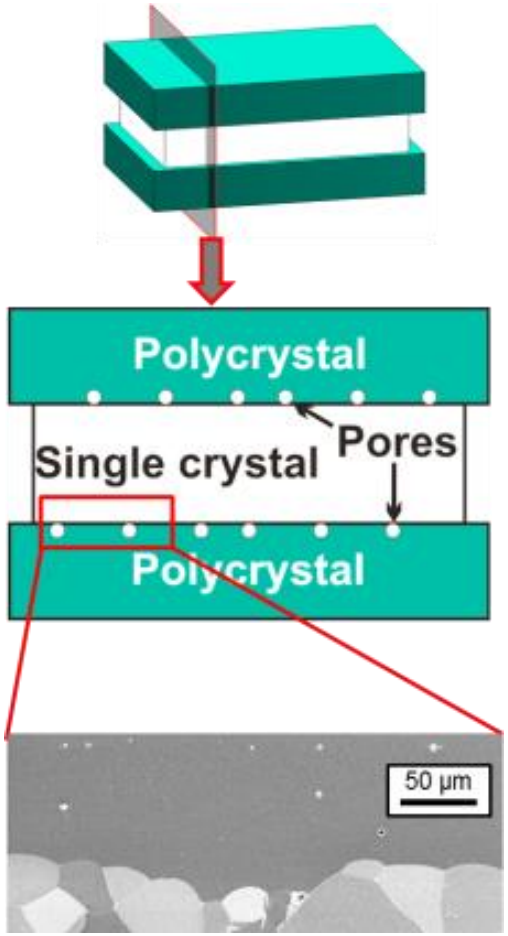
Institute for Applied Materials - Ceramic Materials and Technologies



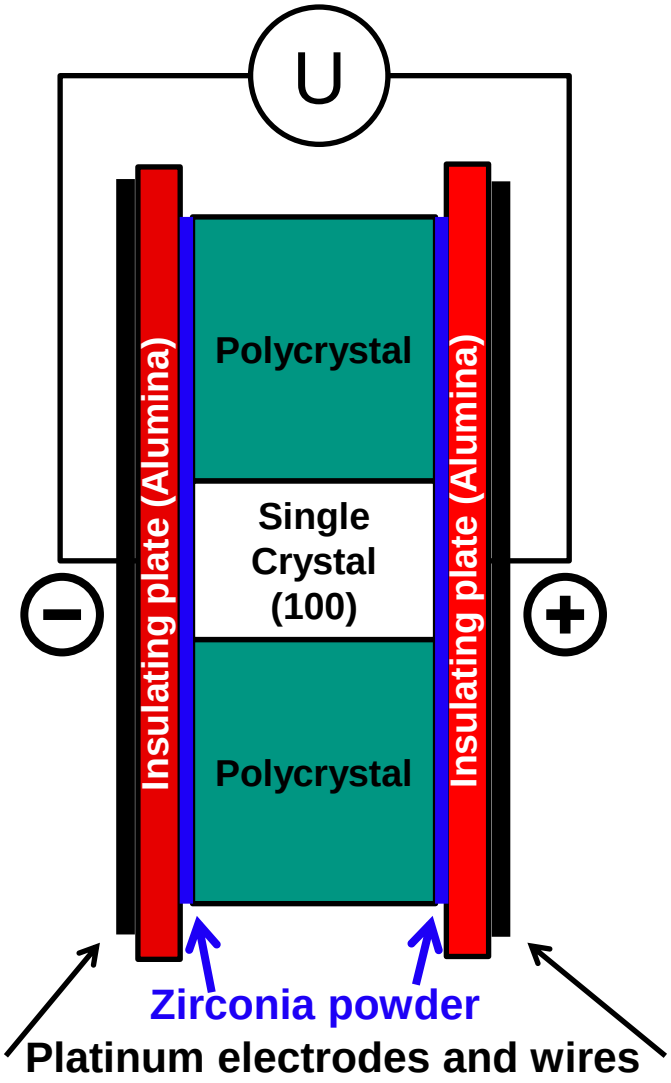
Outline

- Experimental setup
- Growth length of single crystal
- Grain growth in the polycrystal
- Impact of defect chemistry

Experimental setup



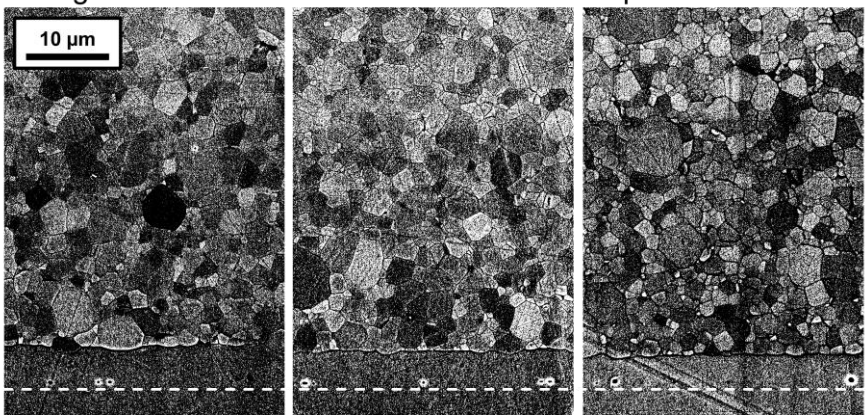
25 V/mm or
50 V/mm



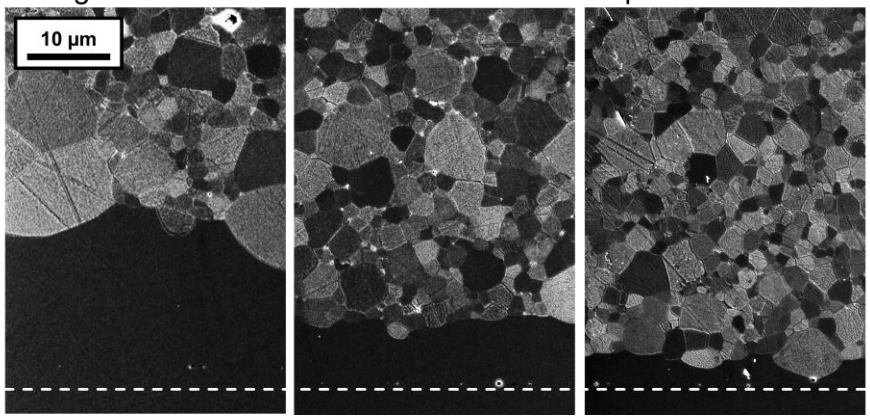
No current due to insulating alumina plates

No chemical reaction due to coarse zirconia powder

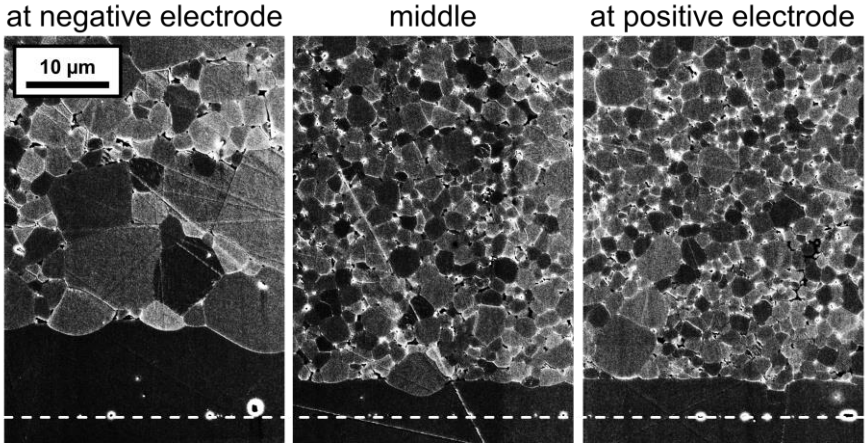
Microstructure for 50 V/mm



1350°C / 10h



1460°C / 2h

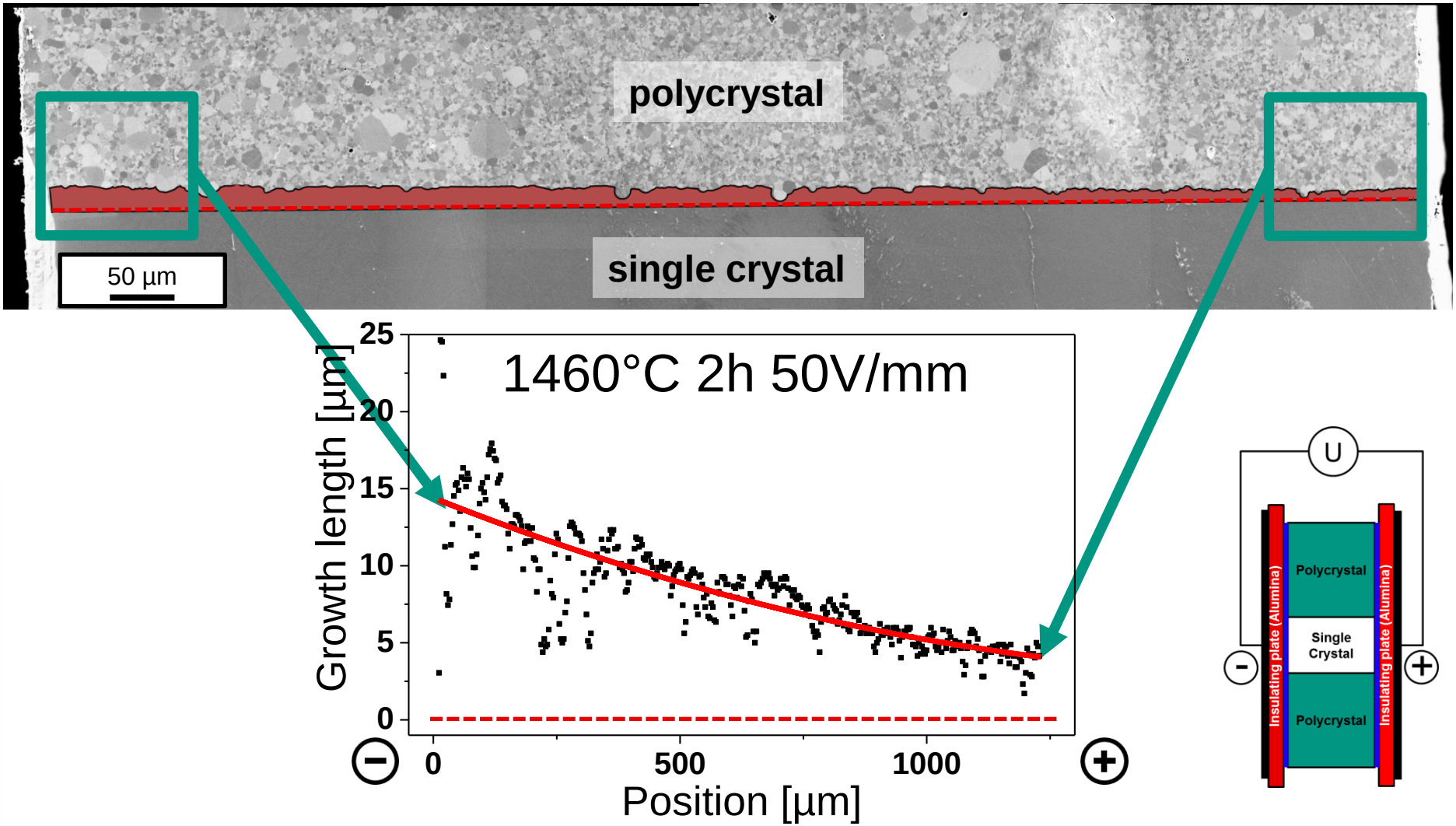


1550°C / 0.5h

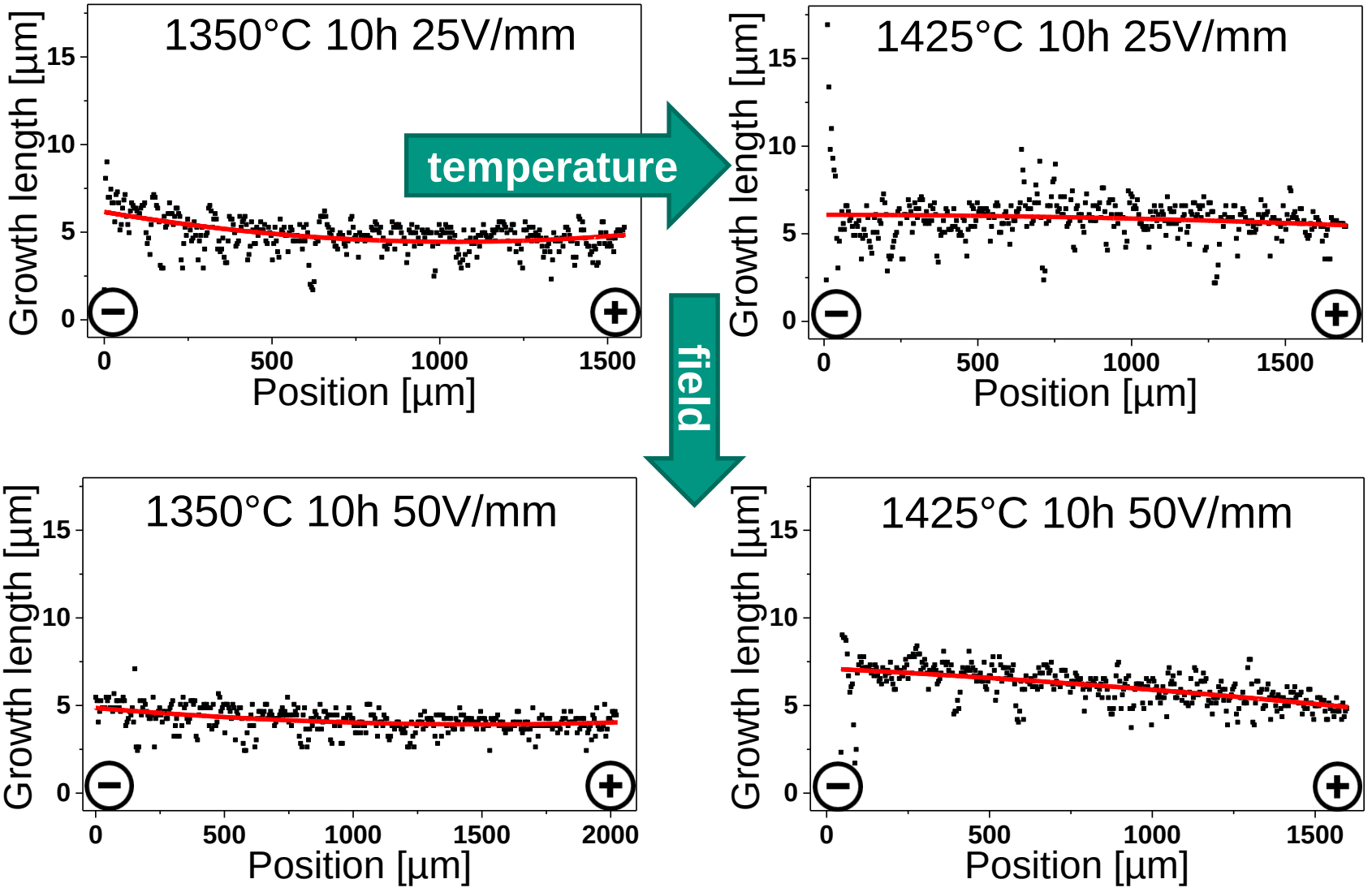
- Information about growth of single and polycrystal
- Impact of an electric field at higher temperatures
- Strong effect at negative electrode

- Experimental setup
- Growth length of single crystal
- Grain growth in the polycrystal
- Impact of defect chemistry

Evaluation of the growth length

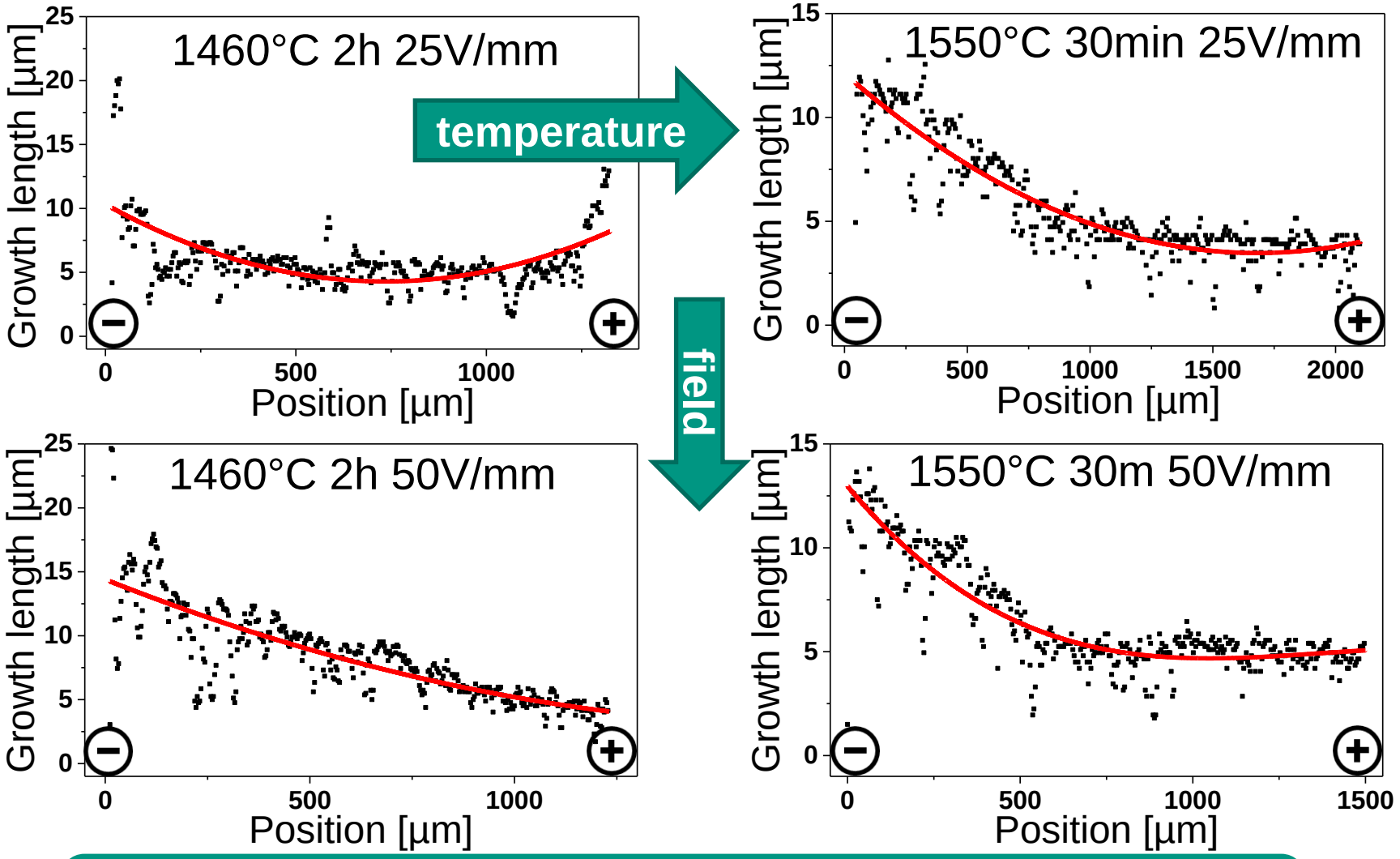


Growth length of single crystals



Weak effect of the electric field up to 1425°C

Growth length of single crystals



Strong effect of the electric field above 1425°C

- Faster growth at the negative electrode & for higher electric field

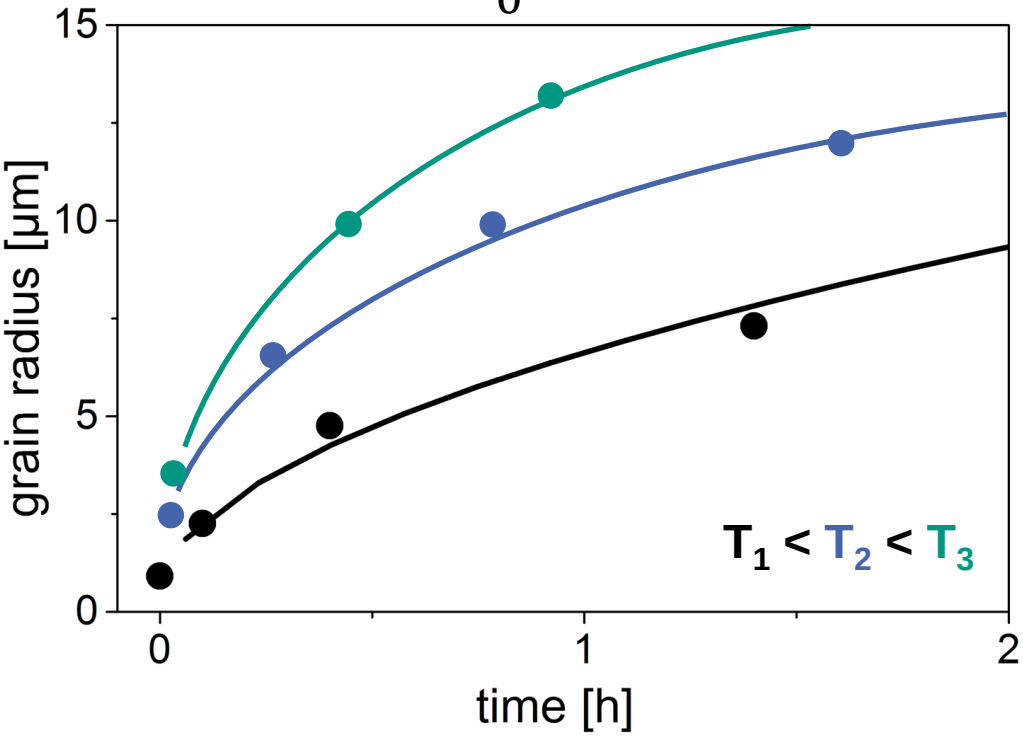
- Experimental setup
- Growth length of single crystal
- Grain growth in the polycrystal
- Impact of defect chemistry

Grain growth in Strontium Titanate

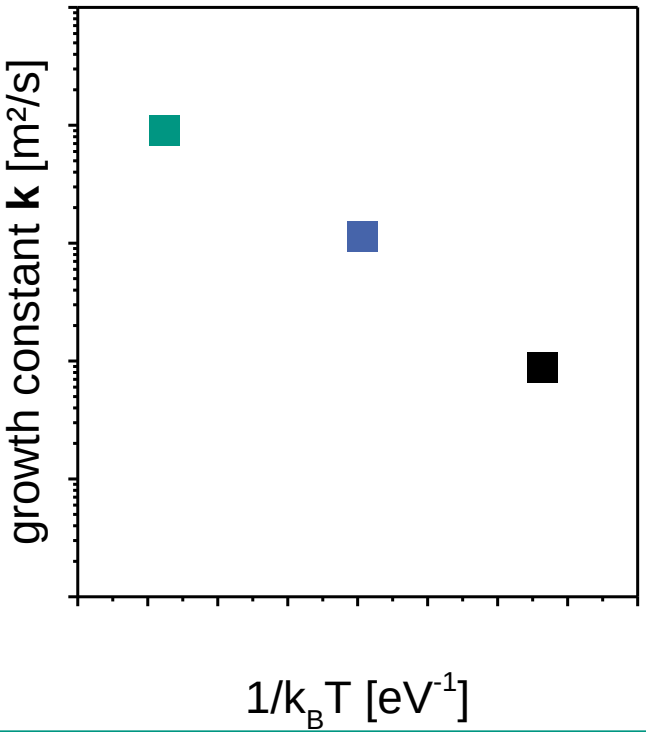
Recrystallization and Grain Growth

Burke, Turnbull, *Prog. Met. Phys. 3*, p.220 (1952)

$$D^2 - D_0^2 = k \cdot t$$

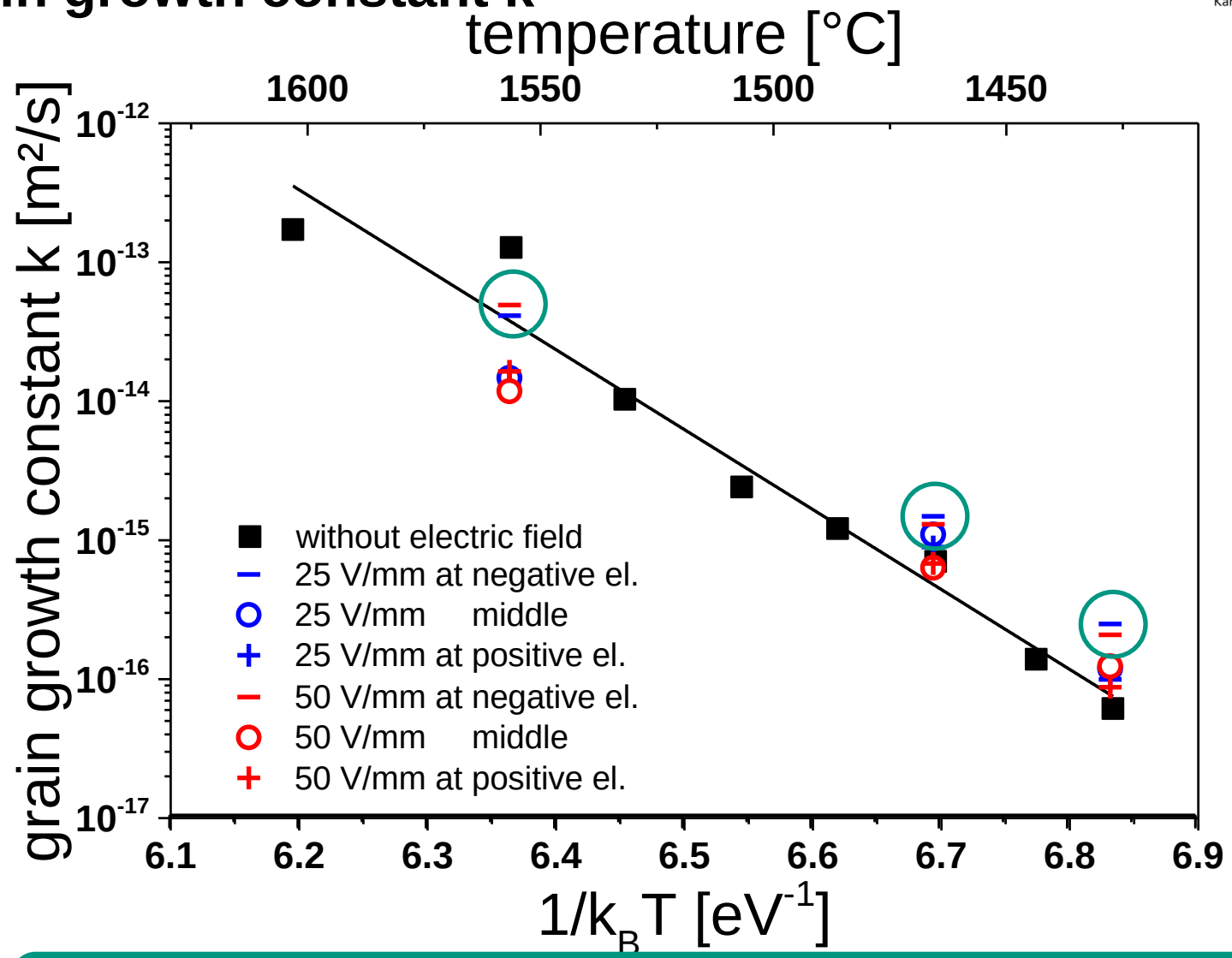


$$k = k_0 \cdot e^{\left(-\frac{E_{GG}}{k_B T}\right)}$$



Grain growth constant k as ‘effective mobility’

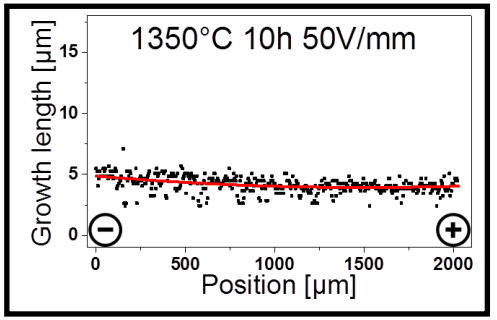
Grain growth constant k



Grain growth overall is similar to results without field !
Strong effect at negative electrode for single and polycrystal !

Grain growth in electric field

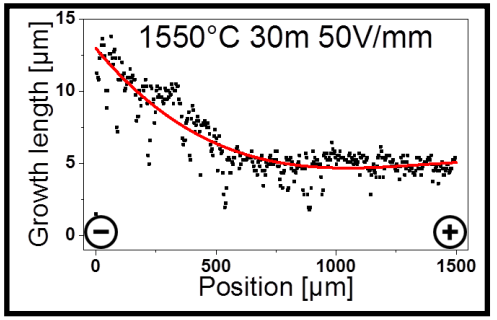
- Up to 1425°C weak impact of an electrical field



- Above 1425°C fast growth at negative electrode

→ Effect is stronger for higher electric field

→ Effect is similar for single and polycrystal

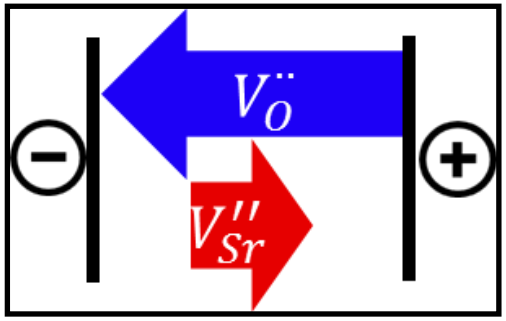


What happens in the material?

→ Defect redistribution

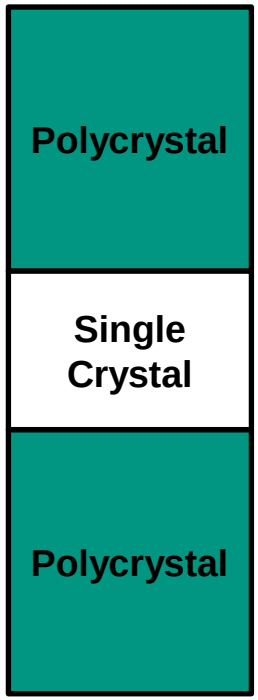
- Oxygen vacancies $D(V_O^{\cdot\cdot}, 1400°C) \approx 10^{-6}\, cm^2/s$
- Strontium vacancies $D(V_{Sr}^{''}, 1400°C) \approx 10^{-14}\, cm^2/s$

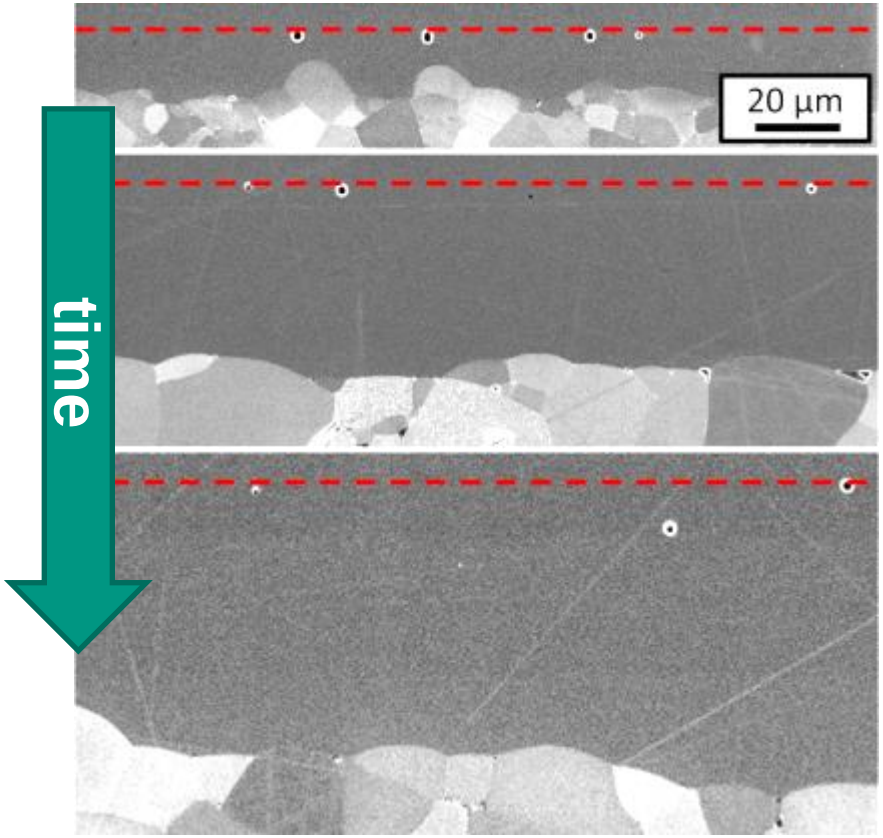
Moos, Härdtl, *J. Am. Ceram. Soc. 80* (1997)
Meyer, Waser, Helmbold, Borchardt, *Phys. Rev. Lett. 90* (2003)



- Experimental setup
- Growth length of single crystal
- Grain growth in the polycrystal
- Impact of defect chemistry

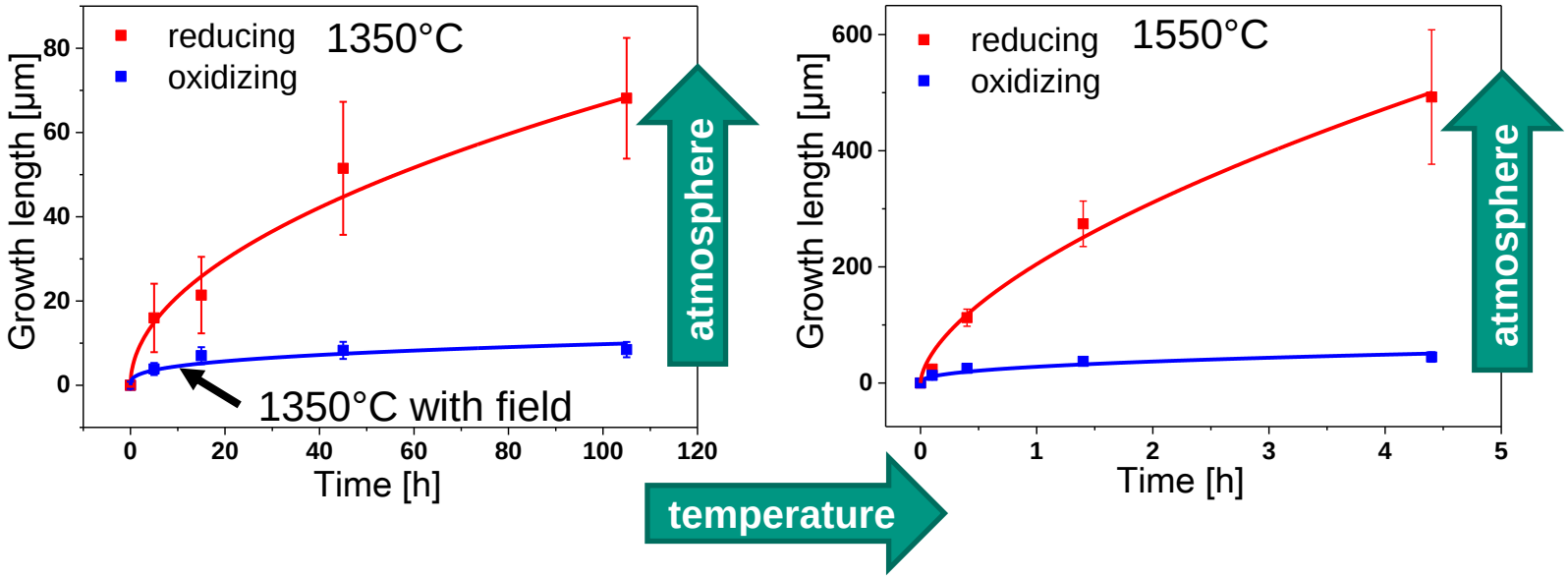
Oxygen vacancies and grain growth





Parameter
- dwell time
- temperature
- **atmosphere**

Oxygen vacancies and grain growth

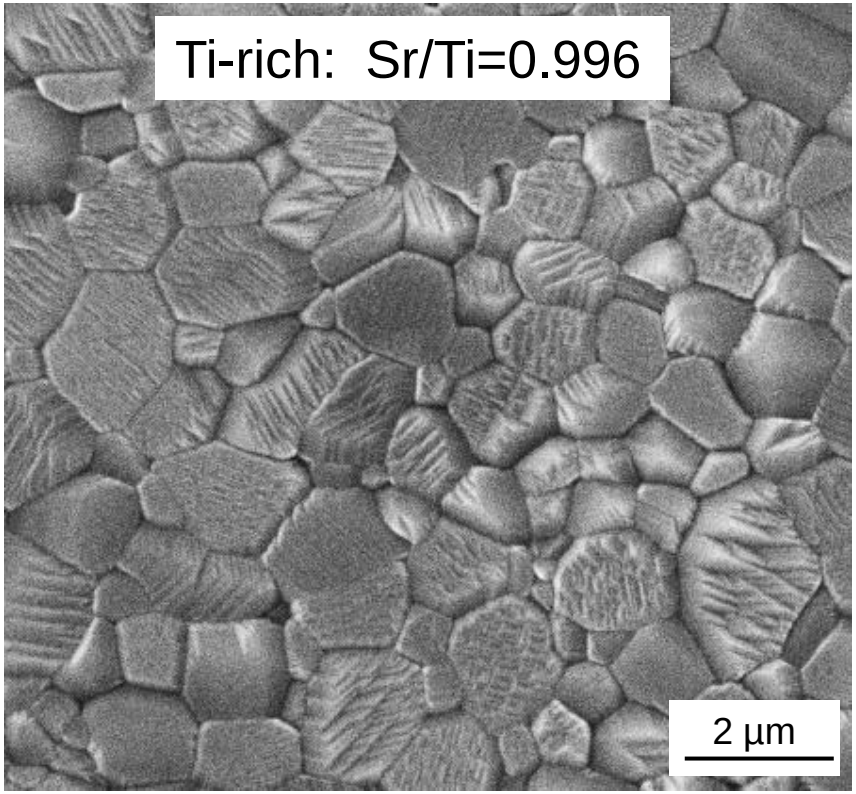


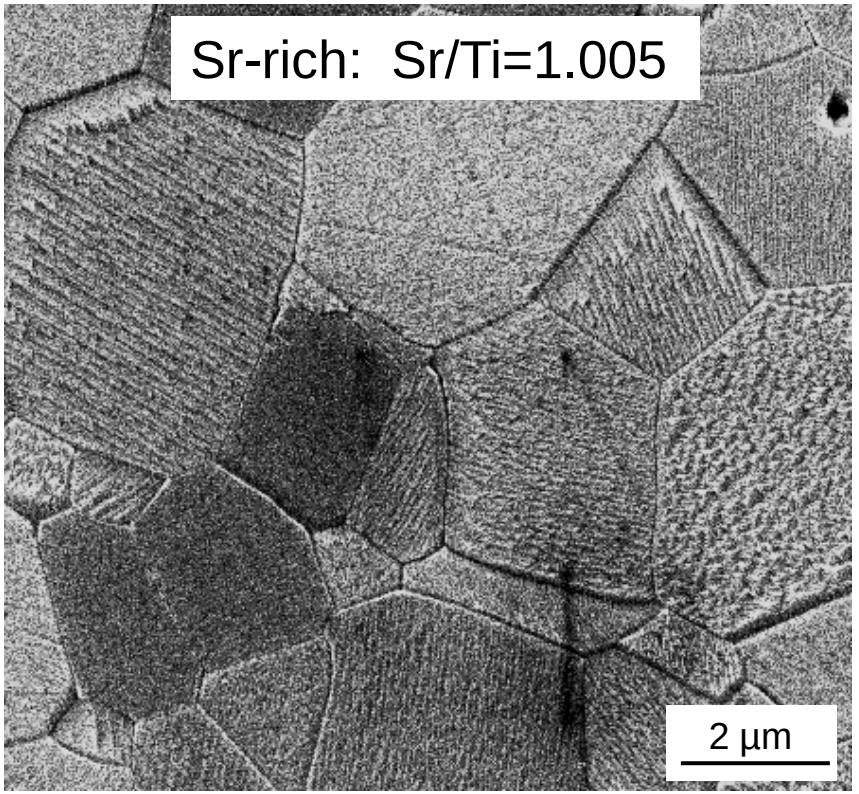
$$O_O \leftrightarrow \boxed{V_O^{\cdot\cdot}} + \frac{1}{2} O_2(g) + 2e'$$

Reducing atmosphere creates more oxygen vacancies.
→ Increase in growth rate of single crystal with $\boldsymbol{high}\ [V_O^{\cdot\cdot}]$

Strontium vacancies and grain growth

1425°C 1h in oxygen



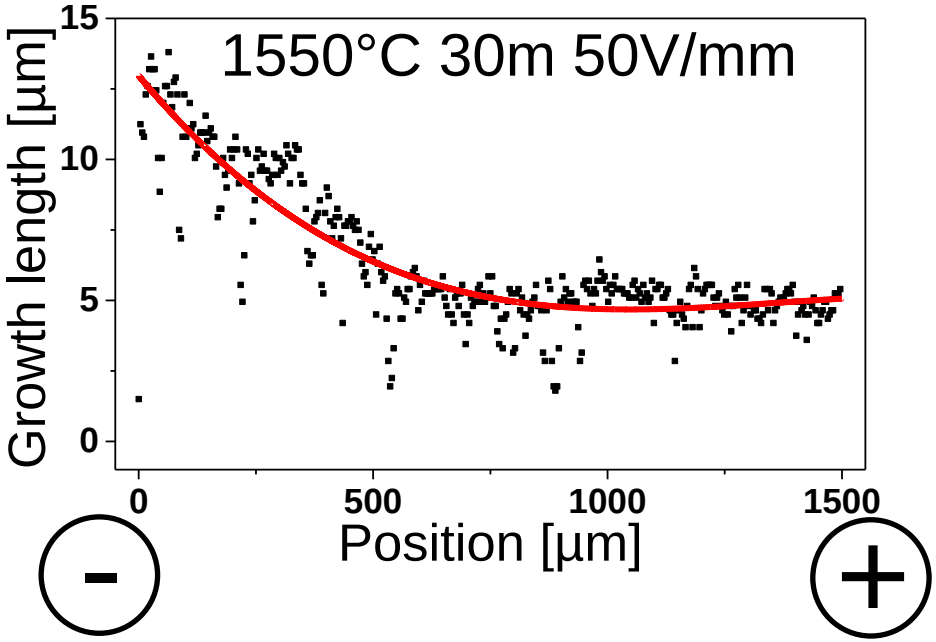


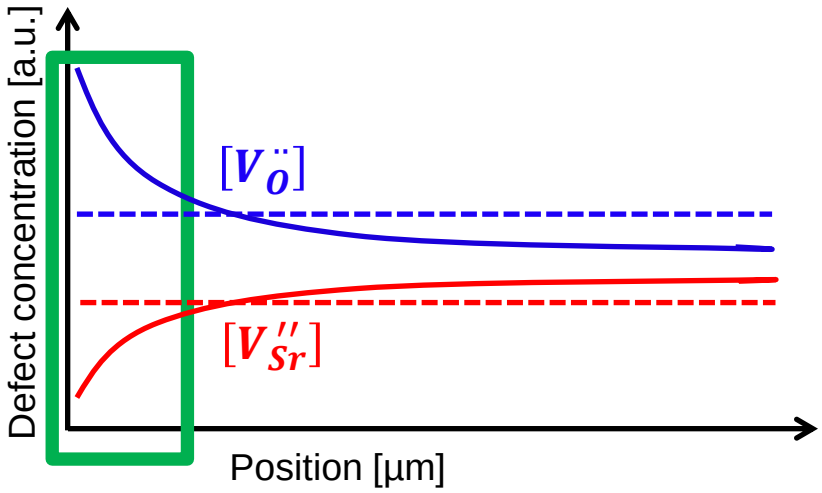
Sr-rich compositions reveal a stronger grain growth than Ti-rich ones.
→ Increase in growth rate of polycrystal with $low\ [V''_{Sr}]$

Effect of electric fields on grain growth

Grain growth is faster, if

- $high\ [V_O^{\cdot\cdot}]$
- $low\ [V_{Sr}^{''}]$





1) **Effect of Sintering Atmosphere on Grain Boundary Segregation and Grain Growth in Niobium-Doped $SrTiO_3$**
Chung, Kang, Vinayak
J. Am. Ceram. Soc. 85, p.2805 (2002)

2) **Effects of donor concentration and oxygen partial pressure on interface morphology and grain growth behavior in $SrTiO_3$**
Chung, Yoon, Kang
Acta Mater. 50, p.3361 (2002)

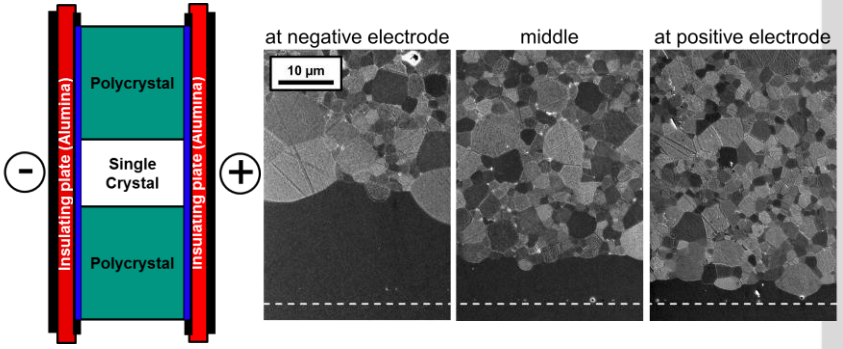
3) **A reversible wetting transition in strontium titanate and its influence on grain growth and the grain boundary mobility**
Rheinheimer, Bäurer, Hoffmann
Acta Mater. 101, p.80 (2015)

4) **Influence of Sr/Ti Stoichiometry on the Densification Behavior of Strontium Titanate**
Bäurer, Kungl, Hoffmann
J. Am. Ceram. Soc. 92, p.601 (2009)

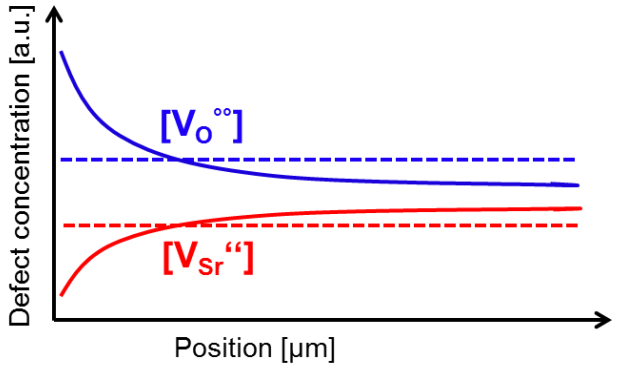
5) **Abnormal grain growth in undoped strontium and barium titanate**
Bäurer, Shih, Bishop, Harmer, Cockayne, Hoffmann
Acta Mater. 58, p.290 (2010)

Summary and conclusions

- Strong impact of the electric field on grain growth in $SrTiO_3$ above 1425°C
  - Appears in both single and polycrystal
  - Faster growth at negative electrode
  - Effect is stronger in higher electric field



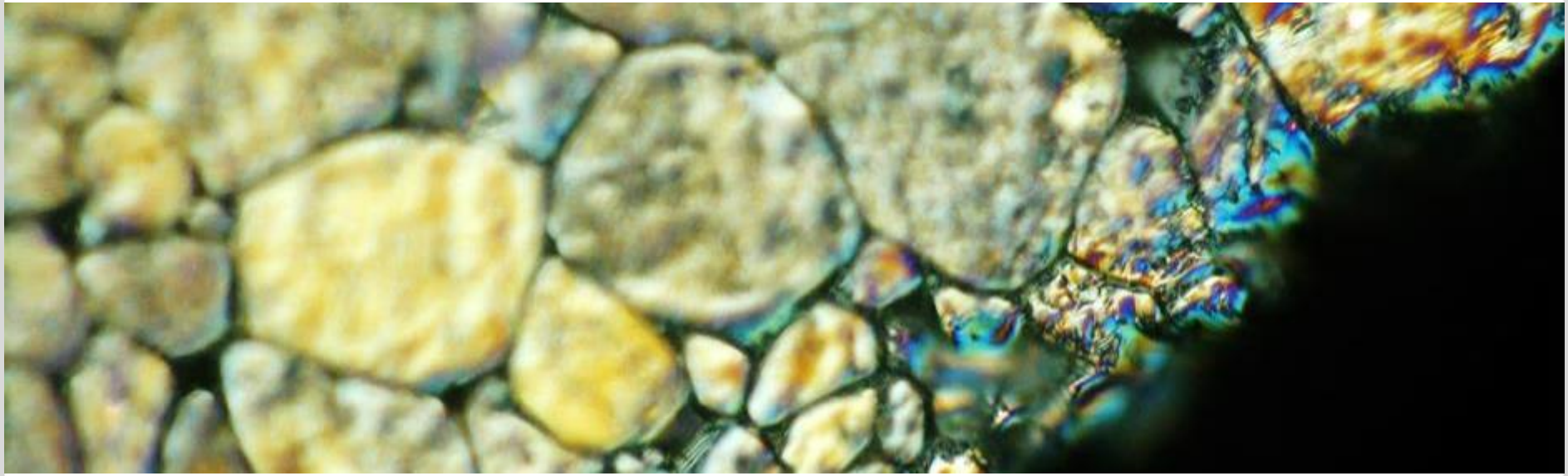
- Fast growth at negative electrode is caused by
  - Redistribution of defects in the electric field
  - High oxygen vacancy concentration
  - Low strontium vacancy concentration



Defects are important for grain growth in perovskites !
Grain growth with and without field can be treated similarly !

Karlsruhe Institute of Technology

Thanks for your attention!



Institute for Applied Materials