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COMPARING GRAIN GROWTH EXPERIMENTS AND SIMULATIONS IN 3D

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ABSTRACT

Experimental investigations in STO ceramics revealed a grain growth anomaly, that presumably originates in the interface anisotropy of these materials. However, the influence of anisotropic grain boundary (GB) properties on grain growth behaviour is to a large extent unknown. The present paper presents 3D grain growth simulations as a possible approach to understand the influence of anisotropic GB properties on the collective behaviour of the GBs in a polycrystal during grain growth. Possibilities to compare 3D growth simulations with experimental 2D sections are discussed and a first reconstructed x-ray diffraction contrast tomography image of a SrTiO₃ specimen is presented.

1. INTRODUCTION

1.1 Grain growth anomaly in SrTiO₃. Perovskite materials find their application in electronic devices such as piezoelectric actuators, sensors or GB barrier layer capacitors. Many macroscopic material properties relevant for these applications are strongly dependent on

grain size originating from the microstructural evolution of the bulk material during sintering. Therefore, the collective GB behaviour at high temperatures is crucial for the design of application-suited perovskites.

Recent experimental investigations on the growth kinetics of SrTiO₃ at sintering temperatures show a strong non-Arrhenius temperature dependency for the effective GB mobility Bäurer, Weygand, Gumbsch, and Hoffmann (2009). SrTiO₃ specimens with various Sr/Ti ratios have been annealed at temperatures between 1100 to 1600 °C and a growth factor k , comprising effective GB mobility and energy was determined by fitting the experimentally obtained grain sizes to the ideal growth law

$$r^2 - r_0^2 = k\Delta t. \quad (1)$$

Here, r marks the grain size, r_0 the initial grain size and Δt is the annealing time. Figure 1 shows the growth factor k as a function of the thermal activation energy for specimens with Sr/Ti ratios in the range of 0.996 to 1.005. For normal grain growth, the effective mobility decreases discontinuously by orders of magnitude between 1300 and 1350 °C and 1390 and 1420 °C, respectively, dividing the observed growth kinetics of SrTiO₃ into 3 distinct growth regimes, thus deviating from the classical Arrhenius law. A decrease in effective mobility

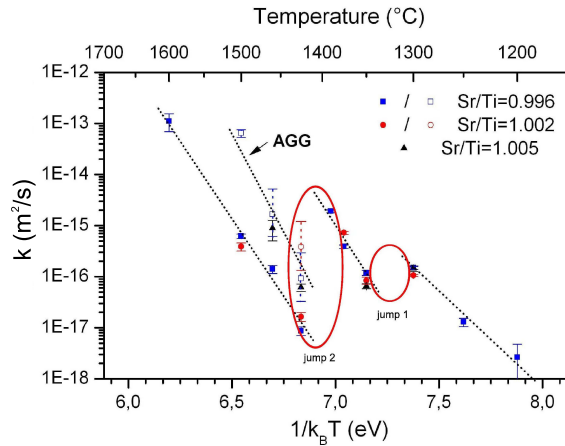


Fig. 1: Effective mobility plotted against the thermal activation energy for specimens with various Sr/Ti ratios, transitions between the different growth regimes are marked in red. In the growth regime marked as 'AGG', abnormal grain growth occurred.

of this size can only be explained by changes in the GB migration mechanism. Although little is known about the collective behaviour of GBs in SrTiO₃ at elevated temperatures, it is likely that changes in the migration mechanism originate in its highly anisotropic Sano, Saylor, and Rohrer (2003) and temperature dependent Lee, Sigle, Kurtz, and Rühle (1999) GB energy.

1.2 Simulation Method. Grain growth simulations are a unique tool to characterize the influence of anisotropic GB energy on the microstructural evolution, since GB properties can easily be adapted and microstructural data is fully accessible in 3D. For the present investigations, the three dimensional vertex dynamics model described in Syha and Weygand (2010), Weygand, Bréchet, Lépinoux, and Gust (1999) was applied. The model is able to handle misorientation and inclination dependent GB properties. In order to model the growth kinetics of SrTiO₃, we used the empirical functional for the surface energy γ of SrTiO₃ at 1400 °C presented in Sano et al. (2003) and calculated the GB energy γ_{GB} as a function of the orientation according to

$$\gamma_{GB}(\vec{n}) = \frac{1}{2} (\gamma(\vec{n}_l) + \gamma(\vec{n}_r)), \quad (2)$$

where \vec{n}_l and \vec{n}_r are the GBs surface normal with respect to the left and right grains local coordinate system respectively.

As can be seen from figure 2 first comparisons of experimentally determined and simulated grain size distributions, model and experiment are in good agreement.

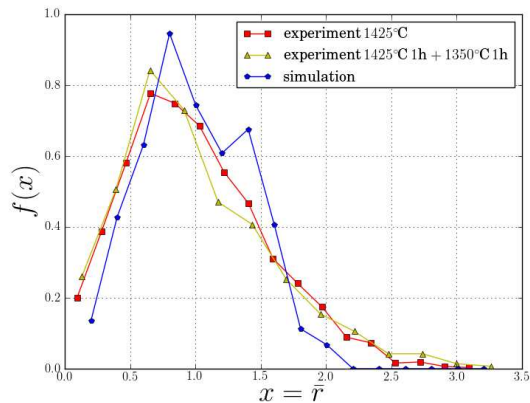


Fig. 2: Grain size distributions resulting from experiments with various annealing parameters and vertex dynamics simulations using the empirical functional for the surface energy of SrTiO₃ at 1400 °C presented in Sano, Saylor, and Rohrer (2003), respectively.

2. RESULTS

2.1 Comparing 2D and 3D data. Comparing evolved structures of simulations with isotropic and anisotropic GB energies, respectively revealed major differences in triple line angle distributions, see figure 3(a). Here, the angle distribution at the triple lines taken from a 3D dataset is shown. While the angles are mostly close to the expected 120° in the isotropic

case, the angle distribution broadens significantly when applying an anisotropic GB energy. Unfortunately this effect is no longer detectable in 2D serial sectioning data originating from the same modeled structure, see figure 3(b).

Conventional metallographic methods however generate 2D datasets only implying a loss in information compared to a full 3D dataset. Thus we searched for observables concerning orientation dependent GB characteristics that allow for a proper comparison of experimental and simulation data. Candidates were the GBs straightness, see figure 4 and the length

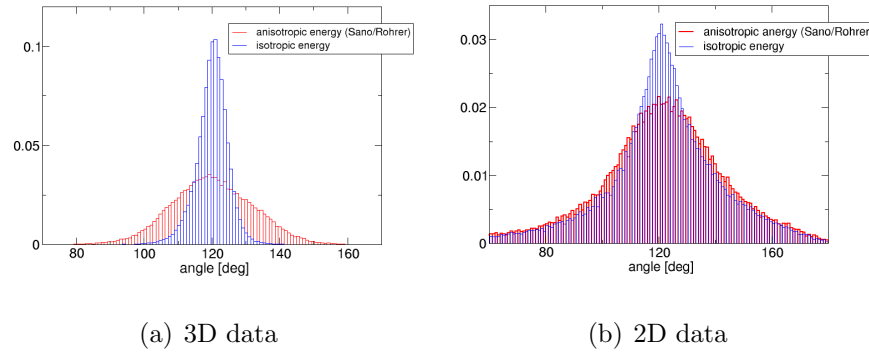


Fig. 3: Angle distributions at the triple lines for isotropic (blue bars) and anisotropic (red bars) simulations. 3D and 2D data were taken from the same modeled structure.

of parallel GB segments, see figure 5. Both observables do not reveal a significant difference between isotropic and anisotropic simulations observed in the 3D dataset. Thus, full 3D microstructure reconstructions of the specimen are needed in order to investigate the influence of the temperature dependent GB energy anisotropy on the microstructure.

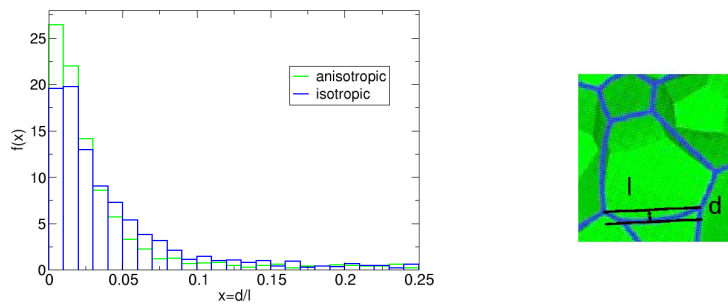


Fig. 4: (a) Curvature distribution for simulations with isotropic (blue bars) and anisotropic (green bars) GB energies, respectively. Data was taken from random sections through a 3D structure. (b) Schematic image of straightness evaluation as $\frac{d}{l}$ in 2D cross-sections of modeled 3D structures.

Comparing grain growth experiments and simulation in 3D

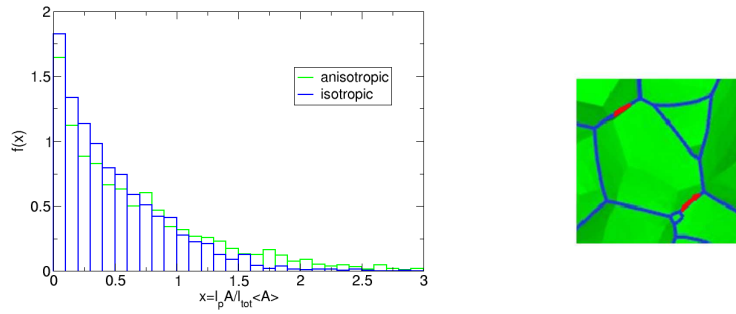


Fig. 5: (a) Distribution of parallel GB segments in random sections through a 3D structure for simulations with isotropic (blue bars) and anisotropic (green bars) GB energies, respectively. (b) Schematic image of parallel GB segment identification in 2D cross-sections of modeled 3D structures.

2.2 Diffraction Contrast Tomography Images. A feasibility study on 3D imaging of perovskites using the X-ray diffraction contrast tomography (DCT) technique described in Ludwig, Reischig, King, Herbig, Lauridsen, Johnson, Marrow, and Buffiere (2009) was carried out at the European Synchrotron Radiation Facility (ESRF) in Grenoble. The data was acquired from a SrTiO_3 specimen with a diameter of $320\mu\text{m}$ using a 40keV x-ray beam. A fully reconstructed grain structure is shown in figure 6. Additional information regarding the crystallographic orientation of the grains is accessible and will be analyzed in future work.

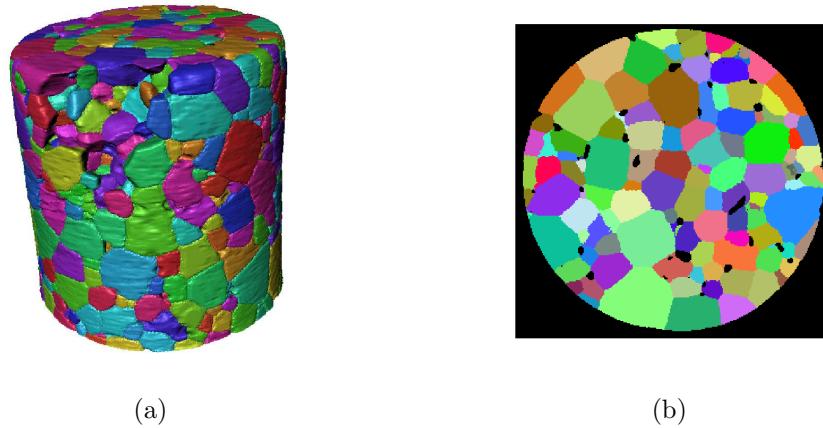


Fig. 6: (a) Reconstructed 3D structure consisting of 1140 grains and (b) 2D section of the cylindrical SrTiO_3 specimen with a diameter of $320\mu\text{m}$. The coloring is according to grain ID, interjacent black regions are porosities resulting from the sintering process.

3. DISCUSSION

While conventional metallographic methods cause a loss in information complicating a comparison between 3D grain growth simulation results and experimental data, the DCT technique allows for a full reconstruction of 3D grain structures, including grain orientations. The present results are the first non-destructive 3D reconstructions of a large ensemble of SrTiO₃ grains. A more systematic application of this technique will allow a study of the grain boundary motion in perovskites more thoroughly and to refine the 3D vertex dynamics model for the simulation of grain growth in these materials.

Further studies combining both, the DCT technique and growth simulations aim at explaining the recently observed grain growth anomaly in SrTiO₃ and to investigate the correlation between grain orientation and morphology in general.

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