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The Structure of Grain Boundaries in Strontium Titanate: Theory, Simulation and Electron Microscopy

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We describe a combination of theoretical and experimental techniques that have been applied to the study of grain boundaries in $SrTiO_3$, with particular attention to $\Sigma 3$- and (100)-oriented grain boundaries.

Several interatomic potentials from the literature have been tested in order to explore how accurately they describe the structures of the $\Sigma 3(111)[\bar{1}10]$ and $\Sigma 3(112)[\bar{1}10]$ boundaries[1]. These potentials are of three types: the rigid ion model, with either fixed formal or partial charges, and the shell model. We have also performed a Density Functional Theory (DFT) study with the CASTEP code[2] on the same boundaries, and used the generated data (interface structures and energies) to evaluate the quality of the interatomic potentials with the GULP code[3]. The different potentials and the DFT calculations all predict two alternative structures for the $\Sigma 3(112)[\bar{1}10]$ boundary, one mirror-symmetric and one displaced by $2a\langle\bar{1}11\rangle/3$, and they predict qualitatively similar patterns of atomic relaxation, but boundary energies, excess volumes and their ordering are not consistent between the models. The energies of mirror-symmetric and displaced $\Sigma 3(112)[\bar{1}10]$ boundaries are indistinguishable within the accuracy of the DFT calculations.

An electron microscopy study, which includes high-resolution transmission and high-angle annular dark field methods, has been applied to mapping atomic columns

and testing the theoretical models. In a near-$\Sigma3(112)[\bar{1}10]$ boundary, prepared from a bicrystal, patches of the symmetric structure are found, with intensities consistent with a deficit of strontium at the boundary plane.

We also test successfully on these boundaries a promising genetic algorithm for discovering low-energy grain boundary structures in an unbiased fashion, including the effect of non-stoichiometry[4].

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