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Ab-initio Calculations on Grain Boundaries of SrTiO<sub>3</sub>

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The understanding of electrical properties of grain boundaries in perovskites is essential for their application to capacitors, varistors and positive-temperature coefficient resistors. The origin of the electrical activity is ubiquitously attributed to the existence of grain boundary donors, usually assumed to be impurities, which set up a double Schottky barrier as they are screened by dopants in the adjacent bulk crystal. But microscopic understanding of the origin of the grain boundary charge has not been provided. It is not known yet if the grain boundary structure itself is the intrinsic origin of the so-called grain boundary donors or whether non-stoichiometry is essential. The relation between atomic structure and electronic properties is studied by combining experiment and ab-initio calculations. The starting structures for theoretical calculations were obtained from Z-contrast images combined with electron energy loss spectroscopy to resolve the dislocation core structures comprising the boundary. Dislocation core reconstructions are typical of all grain boundaries so far observed in this material. They avoid like-ion repulsion, and provide alternative sites for cation occupation in the grain boundaries. Optimized atomic positions are found by total energy calculations. The density of states of each atom at the grain boundaries and in the bulk are compared with experimentally obtained spectra, and the total charge density is used to determine electrical barriers. Calculated differences in vacancy formation energies between the grain boundaries and the bulk determine if vacancy segregation can account for the postulated grain boundary charge.

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