ELECTRICAL AND STRUCTURAL CHARACTERISATION OF A LOW ANGLE GRAIN BOUNDARY IN Fe-DOPED $SrTiO_3$

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Establishing structure–property relationships from the atomic scale upwards is arguably the central theme of materials science. One faces many difficulties, however, in deriving such relationships for grain boundaries in electroceramics, whose overall electrical properties are in many cases determined by the boundaries, on account of the variety and complexity of the boundary structures and chemistry. For the majority of electroceramics the boundaries are found to block the transport of charge and mass; their resistance is frequently attributed to the presence of space charge layers that are depleted of mobile charge carriers (1).

As a material of technological importance, for which the grain boundaries play a crucial role (boundary layer capacitors (2), varistors (3), and sensors (4)); and, from a fundamental point of view, as a model mixed ionic and electronic conductor, whose bulk properties are well understood (5-7), $SrTiO_3$ is one system, whose internal interfaces have attracted much attention. For acceptor-doped compositions, there are several studies of the electrical properties (8-13) and also of the atomistic structure (14-17) of various grain boundaries. Although this body of work has not yielded a comprehensive interfacial structure–property relationship, the first steps in this direction have been made by Leonhardt *et al.* (12), who investigated oxygen mass transport across two different tilt boundaries: a $\Sigma 3$ (111) twin boundary; and a boundary with a misorientation angle of 23.6°, which is close to $\Sigma 13$ (510). They found that the latter boundary significantly blocked mass transport, whereas the structurally *more* perfect twin boundary did not appear to provide any perceptible barrier.

In an attempt to further elucidate the relationship between interfacial structure and electrical properties, we have chosen to study low angle grain boundaries in Fe-doped $SrTiO_3$, as we are then able, in principle, to alter systematically the interfacial structure, and hence the properties, by varying the misorientation angle. In this contribution we present, for the first time, the results of electrical and high resolution structural investigations on a 5.4° [001] symmetrical tilt boundary.

The atomistic structure of the boundary was studied by various Transmission Electron Microscopy (TEM) techniques. Weak beam dark-field imaging revealed that the boundary consists of a periodic array of dislocations; in high resolution TEM the dislocation cores appeared to be amorphous and were separated by regions of strained lattice. The misorientation angle, dislocation spacing and Burgers vector are in excellent agreement with the theory of Read and Shockley for low angle grain boundaries (18).

The electrical properties of the bicrystal were investigated by means of impedance spectroscopy. Measurements were made with YBCO / Ag electrodes over the frequency range 20 Hz < ω < 10^6 Hz in the temperature range 553 K < T < 693 K, and at T = 693 K for 0.01 bar > PO_2 > 0.5 bar.

It was found that the array of dislocations strongly blocks the passage of charge across the interface. Analysis of the impedance data in terms of a double Schottky-barrier model yields a potential barrier height, $\Delta\phi \sim 0.55$ V, which is weakly dependent on temperature and oxygen partial pressure in the investigated regime.

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