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The Atomic-Scale Origins of Grain Boundary Superconducting Properties

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THE ATOMIC-SCALE ORIGINS OF GRAIN BOUNDARY SUPERCONDUCTING PROPERTIES

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Due to the extremely short coherence lengths of the high- T_c superconductors (around 30 Å in the a - b plane), defects such as grain boundaries are obvious barriers to the flow of supercurrent. Within a few months of the discovery of these materials, it was shown how the critical current dropped four orders of magnitude as the grain boundary misorientation increased from zero to 45°. Even today, there is no quantitative understanding of this behavior. A qualitative understanding is however possible through atomic resolution Z-contrast imaging on $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ and SrTiO_3 bicrystal grain boundaries, combined with bond-valence-sum analysis².

The Z-contrast image of a $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ low angle grain boundary in Fig. 1 shows the same kind of reconstructed dislocation cores as seen in SrTiO_3 , containing reconstructions on both the Cu and Y/Ba sublattices. In Fig. 2, an image of an asymmetric 30° boundary in $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ shows the same units and unit sequence as expected for SrTiO_3 . $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ boundaries are wavy because of their non-equilibrium growth process, and therefore mostly asymmetric in nature, although small segments have the symmetric structure as shown in Fig. 3. It seems reasonable to assume that boundaries of other angles will also have similar structures in these two materials.

Atom column coordinates can be extracted from maximum entropy analysis, and bond-valence-sum analysis used to determine the Cu valence in each column. Fig. 4 plots the Cu(1) valence as a function of distance from the grain boundary plane. Although the numerical value in the highly distorted core region cannot be accurate, it is clear that the perturbation to the Cu valence is very significant, and the core region cannot be superconducting. A similar analysis for other angles reveals that the non-superconducting zone increases almost linearly with increasing misorientation angle, explaining the observed exponential drop in critical currents². A similar analysis of a symmetric 36° grain boundary shows a very different behavior (Fig 5). This boundary is constructed from the same cores as seen in Fig. 1, closely spaced and alternating between the two cation sublattices. The Cu valence through the Cu-reconstructed core shows a strong valence drop, but this is *not* seen in the trace across the Y/Ba core.

This would imply that symmetric boundaries have stronger links. The presence of symmetric portions in a predominantly asymmetric boundary (Fig. 3) are likely to be the origin of the strong-linked component deduced recently from transport measurements in magnetic fields³. A 2D schematic of the valence across the asymmetric boundary (Fig. 6) shows a continuous region of reduced valence, with a spectrum of boundary widths due to its quasiperiodic structure. Fig 7 suggests that only a portion of the symmetric boundary has reduced Cu valence.

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FIG. 1.--Z-contrast image of dislocation cores in a low angle $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ grain boundary. Large circles represent Y/Ba columns, small circles Cu-O columns.
 FIG. 2.--Z-contrast image of dislocation cores in an asymmetric 30° $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ grain boundary.
 FIG. 3.--Z-contrast image of a symmetric portion of the 30° $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ grain boundary.
 FIG. 4.--Cu(1) valence across the asymmetric 30° boundary (Fig. 2) showing non-superconducting zone.
 FIG. 5.--Cu(1) valence across Cu and Y/Ba cores in a symmetric 36° boundary.
 FIG. 6.--2D schematic of Cu(1) valence for the asymmetric 30° boundary showing varying widths.
 FIG. 7.--2D schematic of Cu(1) valence deduced for the symmetric 36° boundary.

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