Atomic investigation of Ca segregation at [001] tilt low angle grain boundaries in YbBa$_2$Cu$_3$O$_{7-\delta}$ high temperature superconductor bicrystal thin films

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New constructions and future upgrades of high field magnet devices are highly dependent on high critical current density ($J_c$) high temperature superconductors (HTS) in long length forms, which are inevitably polycrystalline and thus contain lots of grain boundaries (GBs). However, the GBs in HTS are known as an obstacle for carrying supercurrent. Former [001] tilt bicrystal thin film studies showed that $J_c$ across GBs ($J_{c,GB}$) in HTS exponentially decreases as a function of misorientation angle $\theta$ when $\theta$ exceeds 5° [1, 2, 3], demanding complicated and/or expensive processing so that high angle grain boundaries (HAGBs) can be minimized or ultimately eliminated in order to attain high $J_c$. When $\theta$ of [001] tilt GBs is less than 7-8° (so called low angle grain boundaries (LAGBs)), the GB plane is actually considered as an array of GB edge dislocations which are parallel to [001]. Interestingly, with drawback of critical temperature ($T_c$) degradation, Ca addition to the REBa$_2$Cu$_3$O$_{7-\delta}$ (REBCO, where RE denotes rare earth) LAGBs can mitigate the current blocking effects [4]. Previous scanning transmission electron microscope (STEM) studies suggested how Ca affects $J_{c,GB}$ [5, 6]. However its mechanism has not been fully understood. Here we report atomic-structural and stoichiometry studies of Ca-doped YbBCO [001] tilt LAGBs by using an atomic resolution analytical STEM JEOL ARM200cF. Pure and 30% Ca-doped YbBCO bicrystal thin films were grown on SrTiO$_3$ substrates by pulsed laser deposition.

First, we performed line scans of electron energy loss spectroscopy (EELS) across a Ca-doped 6° GB. Figure 1 shows Ca and O concentration variation across the GB dislocation core (the line scan started from A to B in Fig. 1a). As suggested in previous studies [5, 6], strong Ca segregation at the GB dislocation core was observed as judged by the sharp Ca peak at its location (Fig. 1b). Fig. 1c of O concentration variation suggested slight increase of oxygen at the dislocation core as well.

In figure 2, we compare the atomic structure of GB dislocation core in pure and 30% Ca-doped YbBCO. As are seen in fig. 2a and c, the GB dislocation core region seems expanded by Ca-doping. In addition, the Ca-doped dislocation core region shows the darker contrast than the rest of matrix as well as than the pure dislocation core, consistent to Fig.1 that showed segregation of the lighter element Ca. It was thought that Ca segregation occurs so as to relieve the strain around the GB dislocation by Ca substitution to the RE sites. However, comparing the strain map of Fig. 2b and d that were derived from Fig.2a and c by geometrical phase analysis (GPA), respectively, it turned out that Ca segregation did expand both tensile and compressive region around the GB dislocation along the GB plane direction, resulting in narrowing the area fraction of LAGBs where supercurrent can flow. This is a contradictory result because 30% Ca doping increased $J_{c,GB}$. Further details and discussion will be provided at the poster presentation.

References:

Fig. 1 EELS line scan across the Ca-doped 6° tilt low angle grain boundary (LAGB), including the GB dislocation core. (a) Coarse ADF image of the GB with the trace of scanning. (b) (c) Ca and O concentration variation across the LAGB, respectively. Normalized Ca and O content are plotted as a function of distance from A to B in fig. 1a.

Fig. 2 (a) (c) High angle annular dark field (HAADF) scanning transmission electron microscope (STEM) images showing the atomic structure of pure and Ca-doped GB dislocation core, respectively. (b) (d) Distribution of strain field (\( \varepsilon_{xx} \)) associated with a GB dislocation core of (a) and (c) is visualized by using the geometric phase analysis (GPA), respectively. Respective color gradation to blue-black and yellow-white represents compressive and tensile strain. \( \pm 0.2\% \) strain contour is shown as white dotted lines.