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Nucleation and Growth of Single- and Multiple-Domain $YBa_2Cu_3O_x$ Levitators: Influence of Seed Crystallography

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DISCLAIMER

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Abstract -- Large diameter $YBa_2Cu_3O_x$ levitators have been fabricated using seeded melt processing. Two types of levitators have been produced: single-domain, obtained using flat $NdBa_2Cu_3O_x$ seeds, and five-domain, obtained using cubic $NdBa_2Cu_3O_x$ seeds. The difference in these two types of levitators can be attributed to differences in nucleation and solidification processes. In particular, the nucleation of multiple (five) domain levitators may be related directly to the crystallography of the cubic seeding crystals which also exhibit a multiple-domain structure. Single-domain levitators are produced by seeding with plate-like seed crystals which are composed of a single domain. Subsequent growth of the levitator is dominated by anisotropic solidification. Using these types of seeds, we developed a dual-seeded melt textured growth process for the production of bulk bicrystals which are useful for studies of grain boundary transport behavior.

I. INTRODUCTION

Magnetic levitation, trapped field magnets, current leads, and hysteresis motors are among the more promising areas for the use of bulk high temperature superconductors in commercial applications.[1]-[5] In many of these applications, particularly levitation applications, large diameter large-domain samples with strong flux pinning are important in achieving the desired properties, such as magnetic pressure. Directional solidification processing of $YBa_2Cu_3O_x$ (YBCO) with Pt additions has been demonstrated to produce large, bulk samples with strong flux pinning. As a consequence, the production of large domain pellets of $YBa_2Cu_3O_x$ suitable for levitation applications using a melt texture growth process has attracted a great deal of interest.

Seeded melt-textured growth has been shown to be an effective method of producing large-domain samples of YBCO. Seed crystals may be based on $(RE)Ba_2Cu_3O_x$ compounds (RE-123 with RE=Yb, Ho, Y, Dy, Gd, Eu, Sm, Nd) or other materials that are suitably lattice matched to YBCO, such as $SrTiO_3$.[6]-[11] Single crystals of most of the RE-123 materials have been produced almost since the discovery of high temperature superconductivity, most commonly by flux growth techniques.[12]-[15] However, although there is a wide range of RE-123 crystals available, Sm-123 seems to be the most commonly used compound for seeding purposes due to its somewhat higher decomposition temperature.

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In this work, we have used Nd-123 as a seed material for the production of large-domain YBCO levitators. The Nd-123 seeds have an advantage over other RE-123 compounds in that the peritectic decomposition temperature is ≈1068°C which is even higher than the decomposition temperature for Sm-123 (≈1052°C) and therefore allows a somewhat wider range of temperatures to be used during processing. We find that during flux growth of the Nd-123, two different types of crystals are produced depending on the growth conditions. Both types of crystals may be used to seed growth of large-domain pellets. However, the crystallographic structure of the pellet depends on the structure of the seed crystal used. In this work, we have studied the structure of the seed crystals and explored the mechanism by which they form. In addition, we have utilized the single crystal seeds to fabricate bulk bi-crystals of YBCO which we have used to explore grain boundary transport behavior.

II. EXPERIMENTAL PROCEDURES

The Nd-123 crystals are grown by flux growth using a 29 mol. % BaO - 71 mol. % CuO flux to which Nd_2O_3 is added. The powder mixture is pressed into the form of a pellet and placed in an alumina crucible. The sample is then heated in air to a high temperature and held for some time to homogenize the melt, and then slowly cooled to 1000°C. Upon reaching 1000°C, the Nd-123 crystals have formed already and the sample then may be rapidly cooled and the Nd-123 crystals extracted. Depending on the specific processing conditions, either cube-shaped seed crystals or plate-shaped seed crystals are produced. Plate-shaped seeds are produced when the homogenization temperature is 1080°C while cube-shaped seeds are produced when the homogenization temperature is 1100°C. Furthermore, the plate-shaped crystals are frequently found to have nucleated heterogeneously along the crucible wall whereas the cube-shaped crystals nucleate within the melt.

The large domain pellets of YBCO are produced by placing a seed crystal on top of a pressed pellet of the YBCO powder mixture, which contains $YBa_2Cu_3O_x$, Y_2BaCuO_5, and PtO_2. This assembly is then heated in air into the temperature range at which the YBCO powder mixture forms a viscous melt while the Nd-123 crystal remains solid. Upon slow cooling, YBCO nucleates at the seed crystal and grows into the melt. Imposing a small temperature gradient further promotes the growth of large oriented domains into the melt. The processing conditions for the YBCO pellets are the same whether plate-shaped or cube-shaped seeds are used, with the only difference being the seed crystal.

III. RESULTS AND DISCUSSION

The macroscopic appearance of YBCO levitators seeded with cube-shaped seeds or plate-shaped seeds are similar. On the seeded surface, each type of pellet exhibits a surface pattern consisting of a radial line emanating from each corner of the seed crystal. These surface features are attributed to growth ledges rather than any particular domain structure. For example, when a pellet seeded with a plate-shaped seed is polished and examined optically under polarized light, it clearly shows the contrast of a single domain. Further, electron backscattered channeling patterns from the polished surface are consistent with a single-domain structure. In contrast, a polished section of a pellet seeded by a cube-shaped crystal shows a multiple-domain structure, as shown in Fig. 1.

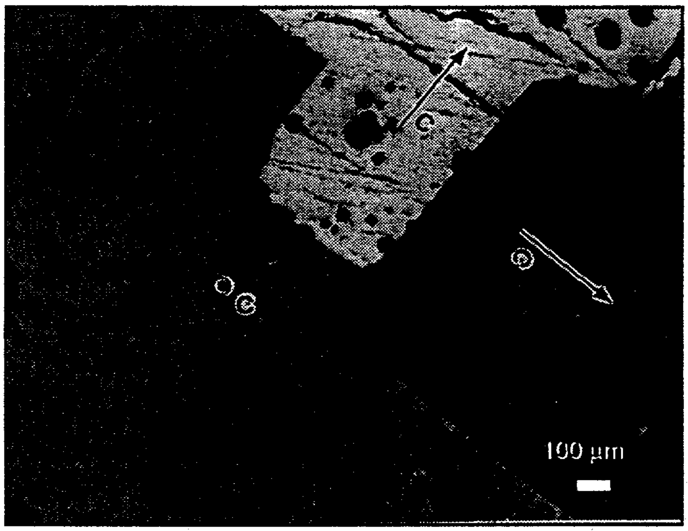

Fig. 1 Optical polarized light micrograph of a YBCO pellet seeded with a cube-shaped Nd-123 seed showing a mulitple-domain structure in the pellet.

Transmission electron microscopy of one of these pellets was used to confirm the multiple-domain structure, as shown in Fig. 2. In this section, portions of two domains are visible and selected area electron diffraction was used to establish the misorientation between the domains as a 90° rotation about [100] as indicated in the figure. Thus, these multiple-domain samples consist of five domains, each of which has nucleated on a different face of the cube-shaped seed crystal with the c-axis normal to the nucleation surface. It should be noted, however, that a single-domain pellet can also be obtained using a cube-shaped seed if the contact area of the pellet is limited to only one face of the seed. In order to exclude the possibility that the multiple-domain structure resulted simply as a result of nucleation at the each surface of the seed, a plate-shaped seed was partly embedded into a pellet prior to processing in order to promote nucleation on more than one surface. In spite of this configuration, a single-domain YBCO pellet was grown.

The formation of multiple-domain samples from the cube-shaped seeds under the same conditions that yield single-domain samples from plate-shaped seeds suggests a difference in the structure between the two types of s eeds. Optical

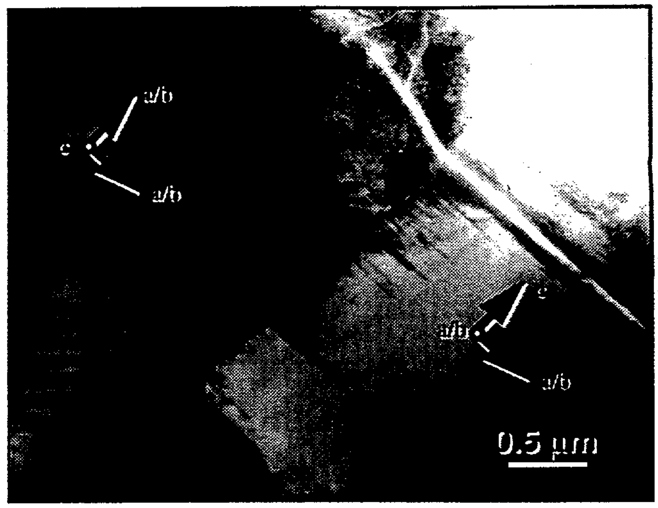

Fig. 2 Bright-field TEM micrograph of a multiple-domain pellet showing the 90° misorientation about [100] between domains.

microscopy, electron backscattered channeling patterns, and x-ray diffraction from plate-shaped seeds all indicate that they consist of a single domain. However, optical microscopy of cube-shaped seeds suggest that they consist of a multiple-domain structure. A comparison of the structure of a plate-shaped seed crystal and a cube-shaped seed crystal is shown in the optical micrographs of Fig. 3.

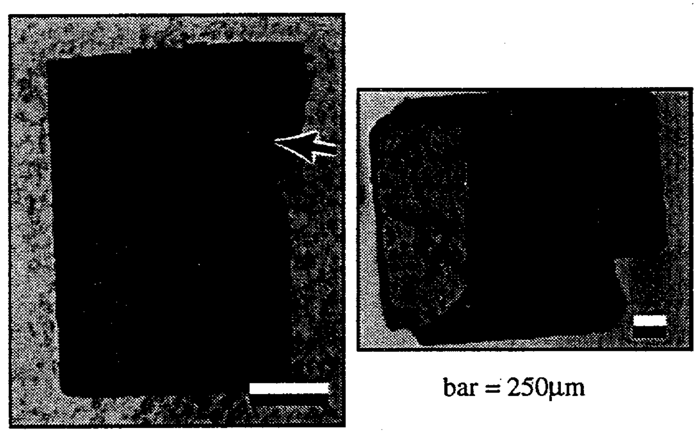

Fig. 3 Optical polarized light micrographs from polished sections of (a) a plate-shaped Nd-123 seed crystal and (b) a cube-shaped Nd-123 seed crystal.

Transmission electron microscopy (TEM) was used to unambiguously establish the domain structure of the cube-shaped seed crystals. A portion of the cross-section of one of these crystals is shown in Fig. 4a. In this image, two domains are visible. The selected area diffraction pattern from a region including both domains is shown in Fig. 4b. Both domains show a [100] zone axis pattern rotated by 90° with respect to one another. Thus, these cube-shaped Nd-123 seed crystals consist of six domains corresponding to each face of the crystal. As in the YBCO pellets, the c-axis in each domain is normal to the surface of the cube face. Thus, we conclude that the multiple-domain structure induced in the YBCO pellets by

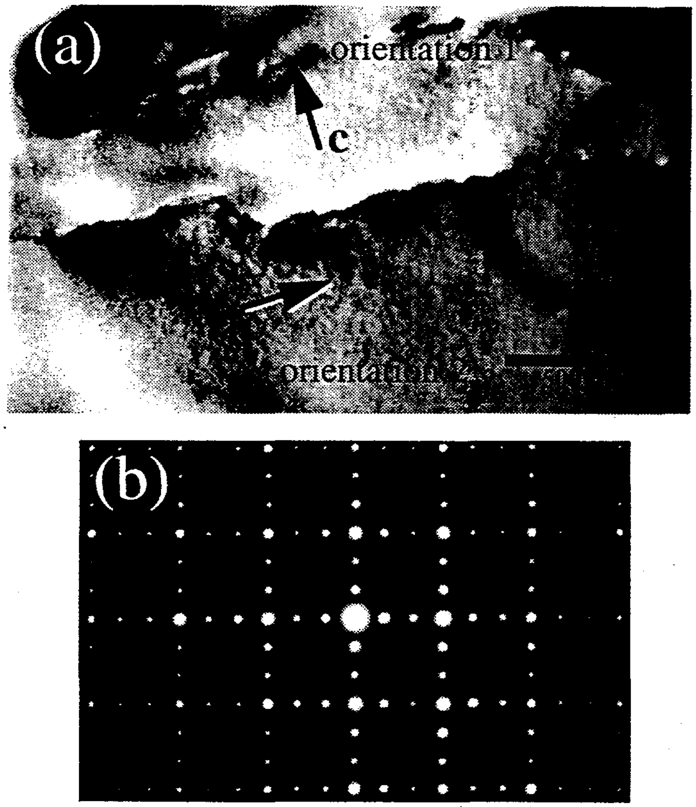

Fig. 4 (a) Bright-field TEM micrograph from a cube-shaped seed showing two domains misoriented from one another by a 90° rotation about [100]. (b) Selected area diffraction pattern showing the 90° misorientation between the two domains shown in (a).

seeding with cube-shaped seeds is a direct result of the multiple-domain structure of the seed itself.

The origin of the two types of Nd-123 seeds with different structures appears to be related to the nucleation of the seeds during flux growth. Both types of seeds show some Al contamination at a level consistent with that reported by others suggesting an interaction of the melt with the alumina crucible. We speculate that the aluminum contamination in the melt plays a role in providing nucleation sites for the cube-shaped crystals. The plate-shaped crystals nucleate primarily at the crucible wall and presumably grow by the leading edge mechanism [16]. In contrast, the cube-shaped crystals nucleate within the melt. Given the higher processing temperature used for growth of the cube-shaped seeds (1100°C compared to 1080°C for the plates), it is likely that a slightly higher concentration of Al may be dissolved into the melt. Thus, it is possible that the amount of Al dissolved into the melt at 1100°C results in a slight supersaturation of Al as the melt is cooled, resulting in the nucleation of $NdAlO_3$. Upon further cooling, Nd-123 may nucleate preferentially on the $NdAlO_3$ crystals. Although we have sectioned a large number of the cube-shaped crystals, we have not yet been able to locate any $NdAlO_3$ at the center of the seed to provide direct evidence of this nucleation mechanism. It is likely that these crystals would be very small and difficult to locate without careful serial sectioning. However, we have generated some less direct evidence that supports this mechanism. One sample was held at the 1100°C processing temperature for an extended time (240 h) and then rapidly quenched by pouring the crucible contents onto a copper block. As shown in the SEM micrograph of Fig. 5, $NdAlO_3$ grains have clearly formed. In addition, although Nd-123 is present in the form of lenticular grains, we believe that this phase did not co-exist with the $NdAlO_3$ at 1100°C but instead formed during the quench. Near the quenched surface where the quench rate was faster, the $NdAlO_3$ grains are also present, but the Nd-123 only appears in the form of dendrites associated with a more rapid solidification process. Thus, we believe the presence of cubic $NdAlO_3$ is responsible for the nucleation of multiple-domain cube-shaped seeds of Nd-123.

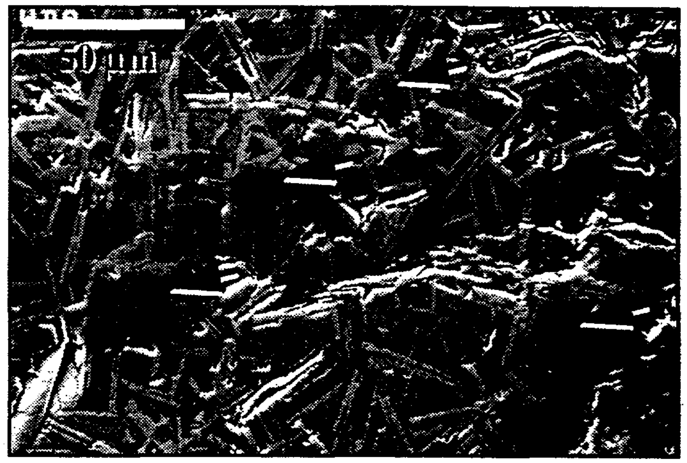

Fig. 5 An SEM image from a polished section of a YBCO pellet mixture quenched after holding at 1100°C for 240 hours. Several $NdAlO_3$ particles which form during this high temperature hold are indicated by arrows.

The ability to grow single- and multiple domain samples from the two different types of seeds offers unique opportunities for additional types of samples to be fabricated and for other studies to be performed. For example, we have used multiple-domain samples to study grain boundary transport behavior across 90° [100] type boundaries of varying tilt and twist character. In addition, we have developed a reproducible technique to prepare bulk bicrystals of YBCO with prescribed misorientations and planar, well defined grain boundary planes [17,18]. The bicrystals are prepared by a dual-seeded melt textured growth process similar to the process used to grow single domain pellets. However, instead of using one single crystal seed, two seeds are placed on top of the YBCO pellet. The bicrystal misorientation is determined by controlling the misorientation between the two Nd-123 single crystals. The grain boundary plane orientation is influenced by the positioning of the seeds and by control of the temperature gradient. Samples for transport critical current measurements can be sectioned from these bicrystals and they exhibit clean, planar grain boundaries on the macroscopic as well as on the microscopic level, in sharp contrast with grain boundaries in thin films that meander [19]-[23]. The cleanliness of the grain boundaries is a consequence of impurity rejection into the liquid ahead of the growth front such that, once the two growing domains have joined and are growing together into the liquid, second phases and impurities are rejected away from the boundary. Using our bicrystals, we have systematically measured the critical current as a function of misorientation angle in bulk samples for the first time. Significantly, due to strong pinning in the bulk material, we were able to measure a difference in the critical current density of nearly two orders of magnitude between the low (≤10°) and the high (≥20°) misorientation angle regime of a series of [001] tilt bicrystals. Furthermore, the critical current density versus misorientation behavior follows the same trend as reported for thin films [24,25]. It is anticipated that the planarity of the grain boundaries produced in these samples may allow a less ambiguous interpretation of the relationship between microstructure and transport properties than is possible from bicrystal thin film boundaries by eliminating the potential variations in properties associated with a varying grain boundary plane associated with meanders in thin film grain boundaries.

IV SUMMARY

In summary, we have used Nd-123 as a seed material for the production of large-domain YBCO levitators. During flux growth of the Nd-123 seeds, two types of crystals may be produced: plate-shaped seeds which consist of a single Nd-123 crystal, and cube-shaped seeds which are composed of six domains of Nd-123 with the c-axis of one domain normal to the surface of each face of the cube. The growth of the two different types of seeds is related to the processing conditions. Both types of crystals may be used to seed growth of large-domain YBCO pellets under identical processing conditions. Seeding with the single-crystal plate-shaped seeds yields single domain YBCO pellets whereas seeding with the multiple-domain seeds yields pellets with the same multiple-domain structure. In addition, we have utilized the single crystal seeds to fabricate bulk bi-crystals of YBCO which we have used to explore grain boundary transport behavior.

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