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AND FLUX PINNING IN MELT-PROCESSED $\text{YBa}_2\text{Cu}_3\text{O}_x^\ddagger$

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Extremely Fine Precipitates and Flux Pinning in Melt-Processed $\text{YBa}_2\text{Cu}_3\text{O}_x$

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The critical current density, J_c , is of utmost importance both in fundamental studies of superconductivity and in practical applications. Impurities and defects can increase the current carrying ability of superconductors. In high- T_c superconductors such as $\text{YBa}_2\text{Cu}_3\text{O}_x$, several defects have been identified, including twin boundaries [1], dislocations [2], oxygen vacancies [3], and Y_2BaCuO_5 (211) [4] precipitates. The phenomenon of 211 pinning, in particular, has generated great interest in the superconductivity community. However, previously reported studies have been controversial about whether 211 particles can act as strong flux-pinning centers. Flux pinning by Y_2BaCuO_5 (211) precipitates was long ago claimed [4], yet some studies conducted by different researchers do not support this conclusion [5,6]. The difficulties encountered in flux pinning studies have resulted from a limited theoretical understanding of flux pinning and a diverse spectrum of complex crystal defects by which the flux lines may be pinned. We have discovered, for the first time, extremely fine Y_2BaCuO_5 (211) precipitates in melt-processed

YBa₂Cu₃O_x (123). From microscopy studies, it was determined that the 211 particles can range from micrometers to approximately 100 Å in melt-processed 123. It has been found that these small precipitates have a high density and finely dispersed in the 123 matrices of the melt-processed samples. It is believed that it is these extremely fine 211 particles (~100 Å) that result in effective flux pinning in YBa₂Cu₃O_x.

In the vortex state of type II superconductors, a supercurrent can flow without resistance in the presence of a high magnetic field below a threshold field: the upper critical field denoted as H_{c2} . Below H_{c2} , as the external field partially penetrates the superconductor, it forms magnetically quantized and elastically flexible flux lines with normal cores. The transport supercurrent interacts with flux lines and generates a so-called Lorentz force, F_L , which tends to push the flux lines away from the equilibrium. Driven by the Lorentz force ($F_L = J \times H_a$, where J is the current density and H_a is the applied field), a phenomenon called "flux flow" will occur, generating an electrical potential in the superconductor. In this case, the material becomes normal as the flux lines "flow" in the superconductor. In a hard type-II superconductor, various crystal defects inevitably exist and can interact with the flux lines, preventing them from flowing. "Flux pinning" is usually the term to describe such a flux line - defect interaction. Obviously, the stronger the flux line - defect interaction, the more pronounced is the pinning effect, and thus the higher the critical current density, J_c . In order to understand the pinning mechanism and to achieve high J_c 's for practical applications, other studies have focused on the relationship between crystal defects and critical current behavior.

For example, Murakami et al. showed that the critical current density, J_c , of the quench-melt-growth-processed (QMGP) 123 has a clear dependence on the density of the 211 particles [4]. They found that as additional 211 precipitates were formed in 123, the J_c increased. Sengupta et al. reported similar results in recent flux-pinning studies [7]. In these studies the 211 particles observed with optical microscopy and scanning electron microscopy (SEM) all exhibit dimensions on the order of 1-10 micrometers, much larger than the coherence length of 123 ($\xi \sim 10 \text{ \AA}$) [4-9]. Research in low-temperature superconductors has shown that a defect cannot effectively pin flux lines unless its dimension is comparable to the coherence length of the superconductor [7]. Therefore, researchers have been uncertain whether these micrometer-size 211 particles can contribute to strong pinning in high temperature superconductor. Moreover, studies by other groups have failed to confirm the benefits of 211 precipitates in melt-textured [5] and zone-melted 123 samples [6]. These discrepancies may have resulted from the great differences in the size of the 211 particles produced by the different processing methods. A recent flux-pinning study [7] showed that enhanced pinning is closely correlated with large amounts of 211 precipitates (on the order of 1-2 micrometers). However, that study compared samples processed by different texturing techniques (i.e., quench-melt-growth and melt-texture -growth) which may lead to some variability in the final microstructures.

Our current study was motivated by the belief that extremely small 211 particles might serve as strong pinning sites. Using a novel processing method [11] we produced melt-textured 123 samples with 211 particles of the order of

100 Å. The method, referred to as solid-liquid-melt-growth processing (SLMG), avoids the high-temperature melting ($>1200\text{ }^{\circ}\text{C}$) required in the case of QMGP [4].

Specifically, we mixed the Y_2O_3 and precursors having the composition of the liquid (L) in the $\text{Y}_2\text{O}_3 + \text{L}$ phase region. The mixture was then melt-processed near $1050\text{ }^{\circ}\text{C}$. To compare the microstructural and flux-pinning differences caused by the extremely fine 211 particles, we prepared two samples; one with a 123 stoichiometry (referred to as the 123 sample) and the other with a 123/211 molar ratio of 5:1 (referred to as the 123/211 sample).

After SLMG processing, we ground the textured samples into powders in a mortar with a methanol bath to form a suspension. Transmission electron microscopy (TEM) experiments were then performed on a Philips CM 30, which was equipped for energy dispersive x-ray analysis (EDX) operating at 300 kV. Under TEM it was obvious that SLMG processing resulted in the formation of very fine inclusions in the 123 matrix. As can be seen in Fig. 1(a) for a representative region in the 123/211 sample, in addition to inclusions as large as 1000 Å , much finer particles are found even of the order of 100 Å in dimension. These are the smallest 211 particles ever reported in bulk 123 specimens. The inclusions are found to be uniformly distributed throughout the sample and have a density of $10^9/\text{cm}^2$. Under TEM examination, the interfaces between these precipitates and the 123 matrix appear to be clean and sharp. No dislocations were observed in the vicinity of the 123/inclusion interfaces. In SLMG processed 123 samples, however, these very fine 211 particles are not observed as shown in Fig. 1(b). It should be pointed out that large densities of micrometer-size 211 have been found in both SLMG processed 123/211 and 123 samples by optical microscopy (11). This observation shows that the particle size in the SLMG-

processed 123/211 samples has a great distribution, ranging from an order of $0.01\text{ }\mu\text{m}$ to $1\text{ }\mu\text{m}$.

Electron diffraction and EDX analysis were also performed on these fine inclusions in the 123/211 sample. From the diffraction pattern, the inclusions were identified as 211. The EDX spectra indicate that the inclusions are rich in yttrium compared with the matrix, thus confirming the 211 identity of these fine inclusions.

To compare the intragranular current densities, we performed the magnetization experiments on finely powdered samples with the average particle size being less than the typical grain size in the textured bulk. (As reported earlier [7], measuring hysteresis curves on powders can reduce the uncertainties in determining critical current density associated with weak-link effects). The samples were crushed with an agate mortar and pestle. The resulting powders were then oxygenated at $450\text{ }^{\circ}\text{C}$ for 24 hours. The average particle size of the powder was about 30 micrometers, as analyzed by SEM. The magnetization measurements were performed in a commercial vibrating sample magnetometer with a superconducting magnet in a gas-flow-type cryostat. The field was ramped at a rate of 80 G/s . For $T > 70\text{ K}$, a platinum resistance thermometer was used; for $T < 70\text{ K}$ a carbon glass type thermometer was used. The powdered sample typically had a mass of about 100 mg.

Figure 2(a) and (b) show the magnetic hysteresis loops at $T = 4.2\text{ K}$ and J_c vs. T at 1 T up to 80 K, respectively, for both samples. The width of the loop, based on the Bean critical-state model [12], is proportional to the flux pinning strength (or J_c). The 123/211 sample with extremely fine 211 particles exhibited

significantly larger hysteresis compared with that of the 123 sample up to 80 K at 1 T. The J_c of the 123/211 samples was found to be significantly higher than that of the 123 sample up to 80 K, as shown in Fig. 2(b), indicating greatly enhanced flux pinning in the 123/211 sample.

Work is under way to understand the exact mechanism of formation of the extremely fine 211 particles during SLMG processing. We hypothesize that these extremely fine 211 precipitates lead to enhanced flux pinning, as observed in the SLMG-processed 123. There is no evidence of stacking faults or dislocations associated with these fine 211 precipitates, as is the case with coarser 211 particles [15]. According to the classical pinning model [13], when either the size of and spacing between the pinning centers is less than the superconducting penetration depth, there will be a induction difference at the pinning sites, causing a so-called core pinning. In the SLMG samples in this study, the diameter of the 211 particles is on the order of $100 \text{ \AA}$, which is much less than the penetration depth of 123 ($\lambda \sim 1000 \text{ \AA}$) [14]. Thus, we conjecture that the great increase of J_c in the 123/211 sample shown in Fig. 2 is likely due to the strong core pinning effect associated with the fine 211 precipitates. However, this conjecture does not exclude the possibility of magnetic pinning [13], since 211 precipitates have a distribution in both size and spacing. In addition, we cannot at this point distinguish between pinning by the fine ($\sim 100 \text{ \AA}$) 211 and the coarser ($\sim 1 \text{ \mu m}$) 211. It is likely that both types of particles enhance pinning, although by different mechanisms.

In conclusion, for the first time a high density of 211 precipitates with dimensions on the order of $100 \text{ \AA}$ have been observed in SLMG-processed 123.

Flux pinning was found to significantly increase up to 80 K, with a large amount of these very fine 211 inclusions.

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Figure captions

Fig. 1 TEM photographs showing (a) SLMG-processed 123/211 consisting of fine 211 particles of the order of $100\text{\AA}$, (b) very fine 211 ($\sim 100\text{\AA}$) particles are not present in the SLMG-processed 123 sample.

Fig. 2 (a) Magnetic hysteresis for SLMG-processed 123 and SLMG processed 123/211 at $T = 4.2\text{ K}$ and (b) J_c vs. T at 1 T for both SLMG processed 123 and 123/211 samples.

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