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Superconducting properties of grain-aligned $HgBa_2CuO_{4+x}$

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$HgBa_2CuO_{4+x}$(Hg 1:2:0:1) particles (15 wt %) were aligned in an applied magnetic field of 70 kG at room temperature in an epoxy resin. Magnetization hysteresis loops were measured from 0 to 10 kG at 20 K for samples oriented with their c axis either parallel or perpendicular to the applied field. The degree of anisotropy, characterized by the ratio of ΔM ($H \| c$ axis) to ΔM ($H \perp c$ axis), was found to be field dependent. A maximum value of 4.53 was observed at 0.5 kG; however, the anisotropy decreased dramatically with increasing field and was isotropic above 3 kG. This behavior differs significantly from grain-aligned $YBa_2Cu_3O_{7-x}$, where the anisotropy is independent of applied field, and may indicate that flux pinning is more isotropic in Hg 1:2:0:1. The critical current density J_c was estimated by the Bean model to be approximately 7×10^4 A/cm^2 at 8 kG, 20 K and was independent of Hg 1:2:0:1 sample orientation.

The recent discovery of superconductivity at 94 K in $HgBa_2CuO_{4+x}$, a single-CuO_2-layer compound, by Putlin *et al.*[1] has generated a great deal of interest in this system. Wagner *et al.*[2] have synthesized $HgBa_2CuO_{4+x}$ (Hg 1:2:0:1) and characterized both its crystal structure and potential defect sites as well as the influence of oxygen on its superconducting properties. Welp *et al.*[3] have measured the irreversibility field of polycrystalline Hg 1:2:0:1 and found its dependence on temperature to be much weaker than that observed for Bi- or Tl-based superconductors. In addition, a value of the irreversibility field of about 0.3 T was measured for Hg 1:2:0:1 at 77 K which is significantly higher than observed for either the two or three layer Bi-Sr-Ca-Cu-O compounds. Finally, Schwartz *et al.*[4] have demonstrated that neutron irradiation of Hg 1:2:0:1 leads to greatly enhanced magnetization critical current densities and an increased irreversibility field relative to unirradiated material. These results, combined with the predicted low anisotropy of its superconducting properties[1,5] based on the relatively small CuO_2 layer spacing of 9.5 Å, make Hg 1:2:0:1 a technology relevant compound. Due to the unavailability of Hg 1:2:0:1 single crystals, grain-aligned Hg 1:2:0:1 samples were prepared to investigate fundamental superconducting properties as a function of crystallographic direction.

Hg 1:2:0:1 was formed from stoichiometric mixtures of HgO and Ba_2CuO_3 as described previously.[2] The as-synthesized Hg 1:2:0:1 was milled in acetone to produce a fine particle size distribution. The median Hg 1:2:0:1 particle size (D_{ave}) was determined by light scattering (Horiba CAPA-700 Particle Size Analyzer, centrifugal sedimention mode) to be approximately 3.7 μm with a maximum particle size of 10 μm. Hg 1:2:0:1 particles (15 wt %) were then dispersed in epoxy and placed in an applied magnetic field of 70 kG under ambient conditions for 4 h. A similar procedure was reported by Farrell *et al.*[6] to produce grain-aligned $YBa_2Cu_3O_{7-x}$ (YBCO) resulting from its intrinsic normal-state magnetic anisotropy and modified by Lewis, Suratwala, and Arndt[7] to produce partial melt processed, grain-aligned YBCO thick films.

X-ray diffraction (XRD) was used to characterize the degree of c-axis texture in the aligned Hg 1:2:0:1 samples, and this was quantified further by θ rocking curve measurements of the (004) peak. Figures 1(a) and 1(b) depict the XRD powder patterns for randomly oriented, as-milled Hg 1:2:0:1 powder and for aligned Hg 1:2:0:1 powder in epoxy, respectively. The (102) reflection, the strongest peak observed in the random powder pattern, is almost absent in Fig. 1(b). The alignment in the Hg 1:2:0:1 sample was calculated from the XRD results us-

ing the following equation: $P_{(004)}=1-\beta$, where $\beta=(I_{102}/I_{004})^{\text{aligned}}/(I_{102}/I_{104})^{\text{unaligned}}$ and I_{102} and I_{004} are the relative intensities of the (102) and (004) peaks, respectively. The value of $P_{(004)}$ was found to be 0.997 and 0.993 for the top and bottom surfaces of the sample, respectively. Rocking curve analysis of the (004) reflection on the top surface of the sample revealed a 1.9° full width at half maximum (FWHM) as shown in the inset of Fig. 1(b), whereas a value of 2.7° (FWHM) was obtained on the bottom surface. Due to limited x-ray penetration depth, rocking curve analysis was also performed on a polished cross section taken through the interior of the sample parallel to the top surface. In this case, an intermediate value of 2.1° (FWHM) was obtained for the (004) peak. Note these values indicate significantly better c-axis alignment than reported previously for magnetically aligned $YBa_2Cu_3O_{7-x}$ particles[8], where FWHM of the (006) reflection was approximately 20°.

Magnetization hysteresis measurements were made in a superconducting quantum interference device for magnetic fields up to 10 kG at 20 K. Figure 2 shows resultant hysteresis loops for aligned Hg 1:2:0:1 oriented either parallel or perpendicular to the applied field. The degree of anisotropy was characterized from a ratio (R) of ΔM ($H\|c$ axis) to ΔM ($H\perp c$ axis) and critical current densities were estimated using the Bean model (i.e., $J_c=20\Delta M/d$, where d was taken to be the median particle diameter of 3.7 μm). The maximum value of this ratio ($R=4.53$) was observed at 0.5 kG; however, this value decreased dramatically with increasing field as shown in Fig. 3. Above 3 kG, the ratio equaled approximately one, indicating isotropic behavior. At 8 kG, J_c was estimated to be approximately 7×10^4 A/cm^2 independent of sample orientation in the applied field. This result agrees with the value reported by Schwartz *et al.*[4] for unirradiated, unaligned Hg 1:2:0:1 measured at 8 kG and 20 K.

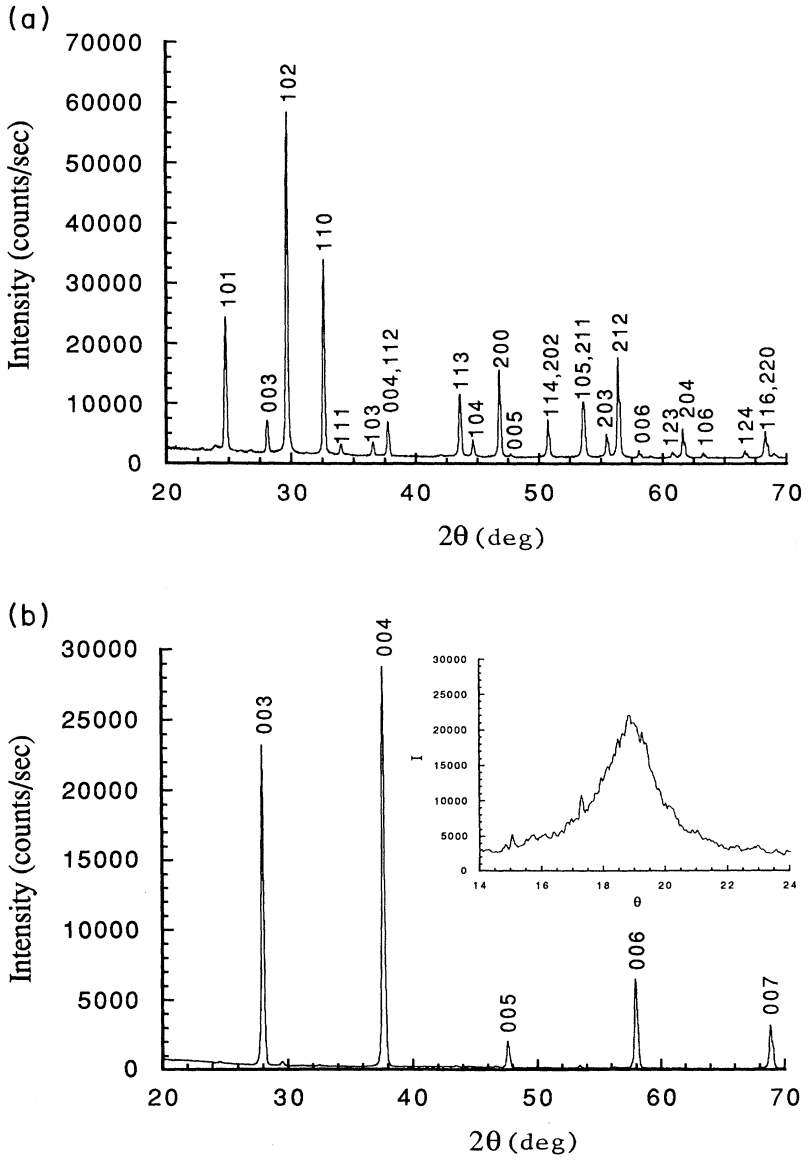

FIG. 1. (a) X-ray-powder-diffraction pattern for randomly oriented, as-milled Hg 1:2:0:1 powder. (b) XRD pattern of aligned Hg 1:2:0:1 powder in epoxy [Note: Inset figure shows the θ rocking curve for the (004) x-ray-diffraction peak.]

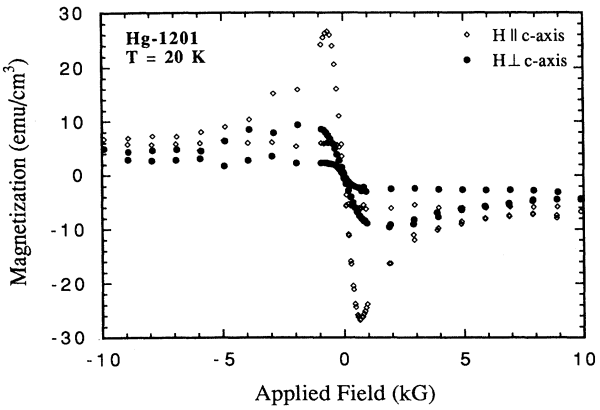

FIG. 2. Magnetic moment hysteresis loops measured at 20 K for the aligned Hg 1:2:0:1. The open circles correspond to $H \| c$-axis, whereas the filled circles correspond to $H \perp c$ axis.

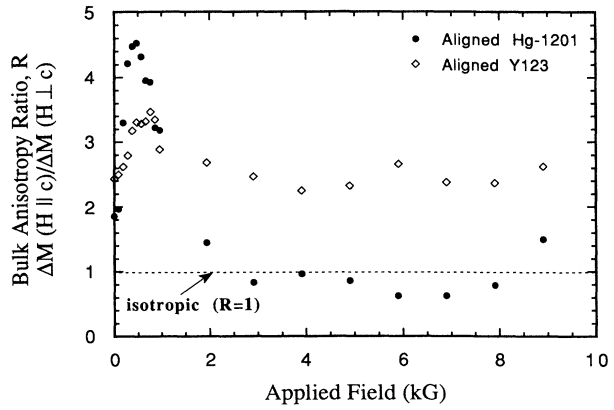

FIG. 3. The dependence of bulk anisotropy (R) on applied field at 20 K for aligned Hg 1:2:0:1 and aligned Y 1:2:3 in epoxy.

The magnetization hysteresis results discussed above differ significantly from those presented previously by Farrell *et al.* for grain-aligned YBCO ($D_{ave} \approx 2-4\ \mu$m) measured at 4.2 K. They observed a significantly higher anisotropy ratio ($R = 10$) that was independent of applied field between 0 kG and 20 kG.[6] The difference in the anisotropy ratio between YBCO and Hg 1:2:0:1 may result from differences in the crystallinity or shape of the starting particles or from intrinsic differences between these materials. In Farrell's work, YBCO particles were formed by mechanically grinding a large YBCO single crystal, whereas the Hg 1:2:0:1 particles used in the present study were formed by milling powder synthesized from a solid-state mixed oxide route. This approach was required to produce the Hg 1:2:0:1 particles due to the unavailability of Hg 1:2:0:1 single crystals. However, this difference in sample synthesis may lead to differences in the crystalline nature or shape of the individual particles. To investigate the first issue, we obtained YBCO powder ($D_{ave} \approx 4\mu$m) synthesized by a similar method as used to produce Hg 1:2:0:1 powder, and prepared aligned samples following the procedure described above for the Hg 1:2:0:1 samples. The degree of c-axis texture in the aligned YBCO was comparable to that observed for the aligned Hg 1:2:0:1, as shown by both XRD [i.e., $P_{006} = 0.982$, where $P_{006} = 1-\beta$ and $\beta = (I_{110}/I_{006})^{\text{aligned}}/(I_{110}/I_{006})^{\text{unaligned}}$] and rocking curve analysis (FWHM = 1.4°) of the (006) peak.[9] Magnetization hysteresis measurements made at 20 K in fields up to 10 kG showed that the anisotropy ratio ($R = 2.7$) was lower than that obtained by Farrell *et al.*, but that it remained essentially independent of applied field after a modest increase between 0–1 kG (refer to Fig. 3). Thus, although the synthesis route of the individual YBCO particles appears to influence the magnitude of the anisotropy ratio, it clearly does not affect the field dependence. To investigate the second issue, scanning electron microscopy was used to evaluate the morphology of the Hg 1:2:0:1 and YBCO powders used in this work.[9] A relatively equiaxed morphology was observed for both powders, which suggests that the observed magnetic anisotropy is not due to differences in particle geometry. Therefore, the difference in the field dependence of the observed anisotropy between these two materials is related to their intrinsic differences, suggesting that flux pinning is more isotropic in Hg 1:2:0:1 relative to the YBCO compound.

In summary, aligned Hg 1:2:0:1 samples were readily fabricated in an applied magnetic field under ambient conditions. Significant differences in the $M(H)$ behavior as a function of sample orientation were observed between aligned Hg 1:2:0:1 and YBCO compounds, even though XRD analysis indicated they had comparable c-axis texture development. The more isotropic nature of flux pinning observed in Hg 1:2:0:1 may be important for several technological applications.

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