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OBSERVATIONS OF VORTEX STRUCTURE IN $\text{YBa}_2\text{Cu}_3\text{O}_7$

BNL--45227

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DE91 000900

Abstract

The remnant magnetic flux distribution in a single crystal and a sintered polycrystal was examined using the magnetic particle decoration technique. The single crystal, which was heavily twinned, showed only local order in the flux lattice. This was probably due to lattice distortions caused by interaction between twin boundaries and fluxons. In the polycrystal, flux pinning effectiveness was seen to vary significantly between grains, depending on size and orientation. Grains with the c axis nearly perpendicular to the applied magnetic field were most effective at pinning flux. Some features of the decoration patterns implied that the flux distribution was influenced by more than just the grains at the sample surface.

Introduction

One of the major challenges in the study of high T_c superconductors is to correlate microstructural features in these ceramics with their macroscopic magnetic and electrical characteristics. To accomplish this, it is necessary to make the flux variations in these materials visible in some way and then examine their distribution on a relatively fine scale. To date, this sort of study has been conducted in one of two ways. The first technique involves examining replicas of the flux distribution at the sample surface, replicas made by depositing ferromagnetic particles on the sample¹. In the second technique, the sample is inspected under polarized light while covered with a layer of transparent material which rotates the polarization vector of light by an amount proportional to the local magnetic field. In this way, light and dark regions corresponding to low and high field become visible in a polarization microscope². The majority of these examinations have been conducted on single crystals with the c axis of the superconductor oriented parallel to the applied magnetic field. They have provided much useful information about the pinning of flux in $\text{YBa}_2\text{Cu}_3\text{O}_7$, and led to the surprising conjecture that some of the regions which do trap flux cannot be associated with any defect or structure visible in the electron microscope³. However, there are still many unanswered questions about flux pinning in this material. There is a further, related materials question which remains unanswered, about what crystal orientation and defect arrangement would result in the greatest flux pinning. In this study, the ferromagnetic particle decoration technique was used to examine flux distributions in single crystal and polycrystalline samples in an attempt to address this issue.

Experimental Details

Two samples were examined in this study, a single crystal and a sintered polycrystal. The single crystal was a platelet prepared at Argonne National Laboratory⁴ using the self flux method⁵. The crystal was approximately 0.1 mm thick, 2 mm in average width, and exhibited many closely spaced twins when examined under polarized light. The sample was marked by several large regions of parallel twin boundaries with a spacing of about 0.5 μm . The polycrystal was produced at Brookhaven by sintering a jet milled Y-Ba-Cu oxide powder with an average particle size of 2 μm ⁶. This powder was pressed into a 2.5 cm diameter disk which was heated in flowing oxygen at 973 °C for 48 h and then slowly cooled. A

Manuscript received September 24, 1990

Figure 1. Schematic diagram of the flux decoration apparatus.

rectangular sample with dimensions 8mm x 3mm x 1mm was then cut from this disk and used in the decoration apparatus. X-ray powder diffraction indicated that the sample was single phase $\text{YBa}_2\text{Cu}_3\text{O}_7$, but transmission electron microscopy revealed nonsuperconducting phases in many of the grain boundaries. Both samples showed superconducting transition temperatures of about 90 K when examined with an AC susceptibility apparatus, though the transition width of the single crystal was smaller than that of the polycrystalline sample.

The decoration apparatus used to mark the flux distribution in these samples is similar to that used by Gammel *et al.*¹. Figure 1

shows a diagram of this apparatus. Tungsten filaments coated with either nickel, iron or gadolinium were electrically heated in a helium atmosphere to produce ferromagnetic particles. These tiny particles drifted downward, where they were then attracted to regions of the superconducting sample which contained magnetic flux. The samples were shaded from the filaments by a baffle, to prevent radiative heating. They had been cooled from 110 K with a magnetic field of 20 Oe applied perpendicular to the sample surface. This applied field had been turned off 10 seconds before decoration, so that only trapped flux remained to create the decoration pattern. The sample was slowly warmed to room temperature in a vacuum after decoration. It was then removed from the apparatus and examined in a scanning electron microscope.

The nature of the evaporated particles depended on the temperature and pressure of the helium atmosphere surrounding the filaments. At very low pressure, polycrystalline films were deposited on the samples. These films exhibited no density changes in the presence of remnant magnetic flux. This is presumably because the superparamagnetic atoms did not interact strongly enough with magnetic field to cause aggregation at fluxons. At higher pressures (>100 Pa) particles with an average dimension of about 10 nm were produced, and these were of large enough moment to be attracted to fluxons. As the helium pressure was increased, particle size generally increased, but this pressure dependence was complex and not fully explored. Above about 600 Pa, the helium column in this apparatus experienced temperature and pressure fluctuations during evaporation which made operation sporadic. Though the particles produced by the hot filaments were quite small, their tendency to agglomerate into threads or clumps while migrating to the sample limited the resolution of the final decoration and obscured detailed positional information about the flux vortices in the superconductor. Clump formation was most pronounced for iron and less of a problem for nickel and gadolinium. Decorations using gadolinium were further complicated by a large spread in particle size and surprisingly low particle production rates. As a result, decorations using nickel have produced the most detailed images.

Examination of a single crystal

The aim of these observations was to correlate the positions of fluxons in single crystal $\text{YBa}_2\text{Cu}_3\text{O}_7$ with defects or other features of the crystal's microstructure. Figure 2 shows a scanning electron micrograph of the surface of the single crystal sample after it had been decorated with nickel particles at 5 K after cooling in an applied field of 20 Oe. There are some signs of clumping in the resultant pattern, perhaps indicating the positions of individual fluxons. However, there is also a good deal of visual noise, scattered nickel particles and strands of particles, which makes the image less easy to interpret. At 20 Oe, the interfluxon spacing should be about 1 μm , and it seems that the clumps do have that approximate spacing. As a comparison, figure 3 shows the surface of the same sample decorated at 100 K without a remnant field, and at six times higher magnification. This pattern shows a homogeneous distribution of particles on a scale much finer than that of figure 2. This indicates that the clumping seen at 5 K is not due to surface effects unrelated to superconductivity. The images of the flux distribution in the single crystal provide no clear evidence for a flux lattice in the material. Though there is some hint of hexagonal or triangular coordination on a local scale, there is no sign of long range ordering in this sample. Attempts to obtain decoration patterns at higher sample temperatures (20 K and 55 K) were largely unsuccessful. These patterns exhibited even more visual noise than the one produced at 5 K, and interpretation was not possible.

There are two possible explanations for the absence of long range order in the flux pattern. Either the flux distribution in this crystal is nearly amorphous, or the flux spacing at 20 Oe is less than the resolution limit of this decoration scheme. The latter is certainly possible, due to the clumping mentioned previously. Since twin boundaries do seem to interact strongly with fluxons³, it is more likely that the twinning in this crystal prevents the formation of a fluxon lattice.

Figure 2. SEM photograph of a single crystal sample decorated with nickel at 5 K in a remnant field of 20 Oe.

Figure 3. SEM photograph of the sample shown in figure 2, decorated at 100 K with no applied field.

Examination of a sintered polycrystal

Virtually all the grains in this sample showed strong preferential growth and exhibited a length to width ratio of greater than 5. About 70% of the area of the sample was made up of small (<100 μm long) grains jumbled together, while the remaining 30% consisted of large, needle-like grains up to a few millimeters long. Figure 4 is an scanning electron micrograph which shows the flux decoration pattern caused by iron particles deposited at 5 K in a remnant field of 20 Oe. After this decoration was made and photographed, the iron particles were removed by swabbing the sample with ethanol. Then, the sample was scanned using a polarized light microscope until the region photographed in the SEM was located. The polarized light micrograph of this region is shown in figure 5. Two of the large grains in these figures have been labeled "A" and "B"

As can be seen in figure 4, the flux distribution in grain A is streaked with regions that have relatively light particle coverage. These regions form parallel bands which run at approximately a 45° angle with respect to the grain boundary. In contrast, the particle distribution on grain B is more uniform, without any noticeable flux free regions. In light of this comparison, it is reasonable to believe that the configuration of grain B is more effective at pinning flux than that of grain A. In figure 5, a cross-hatched pattern of twin boundaries is clearly visible inside grain A, while grain B exhibits parallel twin boundaries running across the grain. The only crystal orientation likely to show the crossed twins of grain A is that with the c axis normal to the sample surface. The angle between the twin boundaries which appear in figure 5 was measured to be within 1° of 90° , further indicating that the basal plane of the crystal is being viewed at normal incidence. The orientation of grain B is not as clearly determined from the optical micrograph. Since there are twin boundaries visible in only one direction, the c axis must make an acute angle relative to the sample surface. If one assumes that twin boundaries occur in both allowed orientations over most of the a-b plane in each grain, as seems to be the case in grain A, then further analysis is possible. Since the twins visible in grain B are all

parallel, this implies that the c direction is in the plane of the sample surface, for in any other orientation, some twin boundaries would converge within the grain.

To further explore the question of how grains with the appearance of grain B are oriented, a portion of the polycrystalline sample was thinned for examination in a transmission electron microscope. One grain in the thinned region appeared to have the needle like geometry and parallel twin boundaries characteristic of grains which seemed to pin flux most effectively. When this sample was tilted approximately 10° with respect to the incident beam, the diffraction pattern of a rectangular lattice with a length to width ratio of 3 appeared. Detailed measurements were not made, so it was not clear whether the a-c or b-c plane had been tilted perpendicular to the incident electron beam. However, for this grain, the c axis was no more than 10° away from the sample surface. Though this information does not prove that the grains with the most densely decorated flux patterns are indeed oriented with the c axis perpendicular to the applied field, that is a likely interpretation of the data. The notion that there are relatively strong pinning centers along the c axis is a reasonable one, since it is known that the superconducting coherence length in that direction is actually smaller than the unit cell of the crystal⁷.

Figure 6 shows a scanning electron micrograph of another grain in the sintered polycrystal. It was taken after decoration of the sample with iron particles at 5 K in a remnant field of 20 Oe. The grain in the center of the micrograph shows a heavy deposit of particles, though due to a shorter evaporation time, the deposit is light in comparison with figure 4. The flux density near the lower grain boundary, marked "L" is less than that at the center of the grain, and the zone of lesser flux density extends nearly $5 \mu\text{m}$ into the grain. The upper grain boundary, marked "U", is partly obscured by iron particles, and the flux density stays high even in the grain boundary. It is as if the decoration has been shifted toward the top of the photograph relative to the underlying superconducting grain. This effect can be seen in several of the needle-like grains, though the direction of the shift is different from grain to grain.

Figure 4. SEM photograph of a sintered polycrystal decorated with iron at 5 K in a remnant field of 20 Oe.

Figure 5. Polarized light photomicrograph of the sample shown in figure 4. Note that labels "A" and "B" refer to the same grains in both figures 4 and 5.

Figure 6. SEM photograph of a sintered polycrystal decorated with iron at 5 K in a remnant field of 20 Oe.

Since the shift is not consistent, it is not likely to be due to a systematic misalignment of the sample holder and magnet; in any case, such a misalignment could be no greater than a few degrees. The feature could be explained if the bulk of these grains extend into the sample at some small angle with respect to the surface. Then, at one boundary, the grain would be quite thin, and perhaps not as effective at restraining flux, so that an undecorated region would appear in the grain near that boundary. Near the other boundary, the strongly pinning grain would be covered with a veneer of the adjacent grain, so that flux trapped in the body of the strongly pinning grain would emerge from the surface in the adjacent grain. In the absence of more detailed three dimensional information about these grains, an authoritative explanation for the decoration pattern displacement noticed in this sample is not possible.

Acknowledgments

The authors would like to thank D. O. Welch for useful discussions in the course of this study, and R. L. Sabatini for his electron micrography. This work was performed under the auspices of the U.S. Department of Energy, Division of Materials Sciences, Office of Energy Storage and Distribution, under Contract No. DE-AC02-76CH00016.

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