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Basic Aspects of High-$T_C$ Grain Boundary Devices

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Abstract: Grain boundaries are extensively used as high-quality Josephson junctions in high-$T_C$ superconductors. Their superconducting characteristics can generally be well described by conventional models of strongly coupled Josephson junctions. Here, we report on highly anomalous critical current vs. magnetic field dependencies of grain boundaries in YBa$_2$Cu$_3$O$_7-x$. Direct imaging with scanning SQUID microscopy provides evidence of magnetic flux generated by single grain boundaries. Conventional Josephson junction models cannot explain these effects if a superconducting order parameter with a pure $s$-wave symmetry is assumed. The results have significant implications for our understanding of the properties of grain boundaries in high-$T_C$ superconductors and for their applications.

1. INTRODUCTION

Single grain boundaries [1] are widely used as high-quality Josephson junctions in a variety of device applications and for experiments to investigate fundamental properties of the high-$T_C$ cuprates, such as the symmetry of their superconducting order parameter [2-4]. In contrast to the textbook behavior of the commonly used $24^\circ$ grain boundary, characteristic features of large-angle grain boundaries are highly anomalous and cannot be explained by conventional Josephson junction properties and by a superconducting order parameter with $s$-wave symmetry. An example of these unusual properties is given by anomalous dependences of the critical current $I_c$ on an applied magnetic field $H_A$. These peculiar characteristics can be accounted for remarkably well, however, if a $d_{x^2-y^2}$ wave component of the order parameter and faceting of the grain boundary are taken into consideration. Based on these considerations, it was expected that single grain boundaries will spontaneously generate magnetic flux [5, 6]. We report evidence of the existence of this self-generated flux, obtained by direct imaging of single grain boundaries with scanning SQUID microscopy (SSM).

2. EXPERIMENTAL TECHNIQUES

The samples used for the experiments were high-quality, c-axis-oriented films of YBa$_2$Cu$_3$O$_7-x$ grown by standard pulsed laser deposition on various bicrystalline SrTiO$_3$ substrates to a thickness of 20–150 nm. A grain boundary we investigated extensively is the asymmetric $45^\circ$ [001] tilt boundary, which is otherwise fabricated by biepitaxy. An image of the surface of such a sample, taken with tapping-mode atomic force microscopy (AFM), is presented in Fig. 1. This micrograph clearly reveals

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Figure 1: AFM image of the surface of a ≈150-nm-thick YBa$_2$Cu$_3$O$_{7-\delta}$ film with an asymmetric $45^\circ$ [001] tilt boundary. The location of the grain boundary is indicated by arrows. The meandering path of the boundary has been replotted at the right of the figure. From Ref. [7].

3. $I_c(H_a)$ CHARACTERISTICS

3.1 Experimental Results

The asymmetric $45^\circ$ boundaries display an anomalous dependence of the critical current $I_c$ on a magnetic field $H_a$ applied in the boundary plane [5, 9-11]. An example of such an $I_c(H_a)$ curve is shown in Fig. 2. Except for a change of the absolute value of $I_c$, these characteristics do not change significantly as a function of temperature or oxygen doping [11]. One of their striking features is the small critical current in zero field $I_c(0)$, which is significantly exceeded at some fields $H_a^* \neq 0$. Such behavior is only consistent with standard junction theories [12] if self-field effects or trapped magnetic flux quanta are present. Both of these effects have to be ruled out in the present case [11].
3.2 Model

The anomalous $I_c(H_a)$ characteristics can be remarkably well explained by a $d_{z^2-\rho^2}$ symmetry component of the superconducting order parameter (see e.g. Ref. [13]) and by the observed grain boundary faceting [6, 10, 11]. For example, in the simplest case, for a $d_{z^2-\rho^2}$ superconductor the density of the supercurrent $J(x)$ flowing across a facet is given by $J(x) = \prod_{m=1,2}((\sin \alpha_m)^2 - (\cos \alpha_m)^2) \sin \phi(x)$ [14], where $\alpha_m (m = 1, 2)$ is the smallest angle between the facet plane and a principal axis of grain $m$, and $\phi(x)$ is the difference of the phases of the order parameters. Concerning the critical current density $J_c(x)$, this implies that for some facet orientations $J_c(x, H_a = 0) < 0$, so that the local current crosses the boundary in the opposite direction of $I_c$ (see Fig. 3). These facets are called $\pi$ facets because for them the grain misorientation causes a shift of $\phi(x)$ by $\pi$. If a field $H_a$ is applied, a gradient $\nabla \phi(x) \propto H_a$ is induced. For some values $H_a^*$ of $H_a$ this leads to $\pi < \phi(x) < 2\pi$, so that $J_c(x, H_a^*) > 0$. Thus, $I_c(H_a^*)$ may be significantly larger than $I_c(0)$. Exact calculations reproduce the measured $I_c(H_a)$ patterns well indeed.

![Figure 2: Critical current as a function of applied magnetic field for an asymmetric 45° [001] tilt boundary in an YBa$_2$Cu$_3$O$_{7-z}$ film at 4.2 K (film thickness: 15 nm, junction width: 16 µm, from Ref. [11]).](image)

![Figure 3: Sketch of the spatial distribution of $I_c$ flowing across a faceted boundary. The arrows indicate the direction of the current across the individual facets.](image)
For zero current bias, a highly interesting behavior has been predicted for the grain boundaries because the phase shift of the $\pi$ facets is expected to induce a Josephson current. If these spontaneous currents exist, they will generate a disordered pattern of unquantized magnetic flux at the boundary [5, 6]. To clarify whether magnetic flux is indeed generated by grain boundaries, we imaged the magnetic fields on the sample surface with high-resolution SSM [15, 16]. In the instrument used [16], a superconducting pickup loop, an integral part of a low-$T_c$ SQUID, is scanned a few microns above the sample surface to image the local magnetic fields. Figure 4 shows an SSM image of a $1024 \times 256 \ \mu m^2$ area of a $45^\circ$ grain boundary imaged with a 4-$\mu m$-diameter pickup loop. The sample was cooled in a field smaller than 1 mG. One bulk vortex pair is visible in the image, as well as spontaneously generated flux in the boundary. This flux is randomly distributed and also changes its sign randomly, with the observed fine-scale variations limited only by the spatial resolution ($\approx 4 \ \mu m$) of the instrument. The characteristic features of this flux are found to be consistent in all respects with the expectations of the model based on the $d_{x^2-y^2}$ wave symmetry component of the superconductor and faceting [17].

Figure 4: Scanning SQUID microscope image of a $1024 \times 256 \ \mu m^2$ area including an asymmetric $45^\circ$ [001] tilt YBa$_2$Cu$_3$O$_{7-x}$ bicrystal grain boundary. The image was taken at 4.2 K without an applied magnetic field after cooling the sample in a magnetically shielded environment (<1 mG). The arrows indicate the location of the boundary. From Ref. [7].

4. IMPLICATIONS OF A $d_{x^2-y^2}$ SYMMETRY COMPONENT ON GRAIN BOUNDARY PROPERTIES AND DEVICE APPLICATIONS

The results described above have several significant implications, both for our understanding of the properties of grain boundaries in high-$T_c$ superconductors and for their applications. A more detailed discussion can be found in Refs. [6] and [17].

- Experimental proof has been presented for the presence of $\pi$ facets at grain boundaries in YBa$_2$Cu$_3$O$_{7-x}$ films. The $\pi$ facets are the most prominent for asymmetric $45^\circ$ boundaries, independent of oxygen concentration and temperature. This provides further evidence of a $d_{x^2-y^2}$ symmetry component of the order parameter in YBa$_2$Cu$_3$O$_{7-x}$.
Transport properties measured in practical grain boundary experiments reflect the averaged behavior of inhomogeneous junctions. The dependence of $J_c$ on grain boundary misorientation, the $I(V)$ characteristic, and the $I_c R_h$ product are not known for the individual facets. Thus, characteristics measured in standard experiments are no direct measure of intrinsic grain boundary properties.

The effects discussed provide a missing link to explain the contradicting results of the symmetry experiments performed by Tsuei et al. [3] on tricrystal rings and by Chaudhari and Lin [2] on biepitaxial polygons. The averaging effect [18] and the presence of self-generated flux will suppress differences of $I_c$ for the various hexagon sites in the biepitaxial samples. If they are taken into account, the results of the polygon experiment become consistent with a $d_{x^2-y^2}$ wave order parameter [17, 18].

$I_c(H_a)$ patterns measured across asymmetric 45° boundaries are a straightforward way to investigate the presence of $\pi$ junctions and possibly also of higher order symmetry components in other high-$T_c$ cuprates.

The magnetic flux is a potential source of noise for grain boundary junctions and for SQUIDs. It can modify the current–phase relation of the junction and enhance critical current fluctuations. Asymmetric 45° boundaries are the worst in this respect.

The flux will cause a paramagnetic component of the grain boundary susceptibility.

Of the three orders of magnitude drop of $J_c$ for an increase of the grain boundary angle from 0° to 45°, about one order of magnitude may be attributed directly to a $d_{x^2-y^2}$ wave symmetry component and to faceting [6].

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