Low-Temperature Anomaly in the Penetration Depth of YBa$_2$Cu$_3$O$_7$ Films: Evidence for Andreev Bound States at Surfaces

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We report the observation of anomalous Meissner currents in thin films of superconducting YBCO with oriented internal surfaces introduced by heavy-ion bombardment. High-precision measurements of the penetration depth ($\lambda$) reveal an upturn in the temperature dependence of $\lambda$ below $T^* \approx 15$ K. The magnitude of the observed effect, its onset temperature and its dependence on the orientation of the surfaces are in quantitative agreement with the theory of surface effects in d-wave superconductors. The anomaly is interpreted to arise from surface currents that are carried by surface-induced Andreev bound states.

The traditional theory of superconductivity in Fermi liquids predicts that superconductors with anisotropic pairing should react sensitively to surface scattering. As a result surfaces are expected to be covered by a layer of thickness of a few coherence lengths of strongly distorted superconductivity with properties different from those of the bulk. These anomalous surface phenomena were first discussed in the context of p-wave pairing in $^3$He [1], later extended to anisotropic pairing in heavy-fermion superconductors [2], and recently to d-wave pairing in cuprate high-T$_c$ superconductors [3-6]. Typical anomalies are suppression of the order parameter ("surface pair-breaking"), zero-bias peaks in the tunneling conductance, anomalous currents flowing in the "wrong" direction relative to the diamagnetic Meissner current, and spontaneous breaking of time reversal symmetry by the surface order parameter $\delta$ [5,7,8]. Recent tunneling experiments [9] report the observation of the zero-bias peak for tunneling into the $ab$-plane, its evolution in a magnetic field, and evidence for a phase transition to a surface state with spontaneously broken time-reversal symmetry. The origin of the surface anomalies is a subtle trapping mechanism of quasiparticles. Quasiparticles which bounce off a surface can be "retro-reflected" back by Andreev scattering, and repeated reflections lead to Andreev bound states. Andreev scattering is sensitive to the anisotropy, phase, and amplitude of the order parameter. Hence, measurements which probe the Andreev bound states may be used to identify the type of superconducting order parameter, both in the bulk and near surfaces.

This Letter presents the first systematic experimental study of the "anomalous Meissner current" (AMC), together with a theoretical interpretation of the data. The AMC is carried by Andreev bound states, and has, in addition to its reversed flow direction, the following signatures. It is confined to a distance of a few coherence lengths ($\xi_0 \approx 15$ Å in YBCO) from the surface, and is therefore a small correction to the total screening current, which is in usual experiments dominated by the regular diamagnetic Meissner current (DMC) flowing within a penetration depth ($\lambda \approx 1500$ Å in YBCO). The scale for the temperature dependence of the AMC is set by the splitting, $\delta$, of the Andreev bound states, and not by the average gap, $\Delta$, which sets the temperature scale for the DMC. One expects $\delta \ll \Delta$, and therefore a characteristically different temperature dependence of the anomalous and regular Meissner currents. In addition, the magnitude of the AMC depends in a characteristic way on the orientation of the surface with respect to the order parameter. All of these signatures were observed in the experiments described below. We interpret these results as evidence for an AMC caused by Andreev bound states. Since Andreev scattering and Andreev bound states are unique features of the Fermi liquid theory of superconductivity we also consider these observations to be strong evidence in support of the Fermi-liquid theory for superconductivity with a d$_{x^2-y^2}$ order parameter in cuprate high-T$_c$ superconductors.

The purpose of our experiments was to study the thin layer of strongly distorted superconductivity which is predicted to cover surfaces of d$_{x^2-y^2}$ superconductor, and to measure the sensitivity of these effects to the orientation of the surface. A direct way to detect the surface anomalies associated with Andreev bound states is to measure the low temperature penetration depth of a sample containing a high density of oriented boundaries. The small size of the surface layers, of order $\xi_0$, requires the preparation of sharp surfaces on the scale of the coherence length, which poses major experimental difficulties. Photolithographic or related patterning techniques are ruled out since degradation at the edges limits the resolution to at best 10 nm. Alternatives, based on artificial grain boundaries such as step edge or bicrystal junctions have orientation problems. Strong faceting on a nanometer scale leads to averaging over many directions and renders the orientation dependent signatures unobservable [13,14]. For these reasons we pursued an unconventional approach, and used ion tracks generated by high-energy heavy-ion bombardment to introduce artificial interfaces [15]. We used epitaxial YBCO films with the c-axis direction perpendicular to the substrate surface (twin size about 100nm from TEM studies). YBCO films were simultaneously grown on batches of eight $20 \times 20 \times 0.5$ mm$^3$ YSZ (100) substrates by thermal coevaporation [16]. The films were covered by 10 nm thick Y$_2$O$_3$ layers in situ to
The magnetic penetration depth, or more generally, the magnetic field response, is determined by a mutual inductance technique described in detail in Ref. [18]. The YBCO film sample is placed between flat pickup and excitation coils operated at 30 kHz. The excitation field, \( B \approx 20-40 \mu T \), is much smaller than \( B_{c1} \), so that the film remains in the Meissner state throughout the measurement. The ratio of the induced voltages in the pickup coil below and above the superconducting transition gives the inverse screening factor \( S^{-1}(T) \) which is a monotonic function of the London penetration depth \( \lambda(T) \). For simple coil geometries (e.g., circular windings) the absolute value of the penetration depth can be calculated exactly by solving Maxwell’s and London’s equations with the usual magnetic field boundary conditions at an interface [18]. The large superconducting area and the flat coil geometry minimizes crosstalk between the coils. The precision for detecting changes in \( \lambda \) is better than 0.5 Å. To adapt the experiment to the irradiation geometry with parallel tracks along specified directions we used elongated racetrack coils instead of circular windings. These coils consist of straight sections of wire which were aligned parallel to the ion tracks as indicated in Fig. 1c. Since the screening current is a mirror image of the driving current it is also driven parallel to the tracks within the undamaged superconductor and without intersecting and averaging material with different degrees of radiation damage. Because only parallel sections of the coils overlap the irradiated window on both ends, the return currents perpendicular to the beam direction are flowing through the non-irradiated film underneath the AuO\(_x\) layer. Since calculations depend on homogeneous film properties the screening factor \( S(\lambda) \) cannot be converted quantitatively into a penetration depth value. However, for small variations of \( \lambda \), i.e., in the low temperature regime, the temperature dependences of \( \lambda(T) \) and \( S^{-1}(T) \) are the same except for a proportionality factor of order unity.

The overall behavior of the penetration depth below 25 K is depicted in Fig. 2. To compare different samples the data have been normalized at 18 K. The choice of the normalization temperature does not play a role for the interpretation of the low temperature effects, as long as the normalization point is \( \approx 16 \) K. Each data set represents 3 or 4 different samples irradiated in the same direction. In Fig. 2, we compare the temperature dependence of the penetration depth of films prior to irradiation with films irradiated at 0°, 22.5°, and 45° with respect to the axis of maximum order parameter (cf. Fig. 1b). The results can be summarized as follows: above 16 K all the samples behave identically, demonstrating the excellent reproducibility of the experiment. However, in the low temperature regime below 15 K the traces split and the penetration depth goes through a minimum at 45°. For the low temperature regime, the temperature dependences of \( \lambda(T) \) and \( S^{-1}(T) \) are consistent with the applied dose of \( 10^{11} \) ions/cm\(^2\). Although a large volume fraction of the YBCO film is radiation damaged, the inductively measured transition temperatures drop by only 2–3 K, suggesting that the residual crystalline material has maintained its original properties.
increase of the anomaly nearly proportional to the density of tracks, and the upturn in the penetration depth reached values of over 1%. An even stronger upturn has been observed in YBCO powder samples previously and shown to depend directly on the surface structure and chemistry of the grains [19]. The authors suggest magnetic surface layers as one plausible physical interpretation. This interpretation in terms of paramagnetic moments can be ruled out in our case by the characteristic dependence of the anomalies on the irradiation direction. Different samples irradiated in the same direction show the same anomaly, whereas the absolute magnitude of the penetration depth scatter from sample to sample uncorrelated with the direction of irradiation. The anomaly caused by paramagnetic moments would be correlated with the size of the magnetic field, i.e. with the magnitude of the penetration depth. The existence of the anomaly and its strong correlation with the direction and density of the artificially produced boundaries can also not be explained by the standard theory of s-wave superconductivity. On the other hand, the d-wave model of superconductivity provides a natural interpretation of the anomalous Meissner currents in terms of current carrying Andreev bound states (ABS) at surfaces. These states are coherent mixtures of electrons and holes on the Fermi surface, and can be classified for clean superconductors and perfect surfaces by their momentum parallel to the surface. Two states with momenta $\pm \mathbf{p}_f$ carry opposite currents and have the same energy in systems with time reversal symmetry. Hence, they are equally populated in thermal equilibrium and carry no net current. Time-reversal breaking perturbations such as a magnetic field split the energies of these states resulting in an uneven population and a net equilibrium current. Of particular importance for the anomalous Meissner currents are zero-energy bound states associated with the sign changes of a d-wave order parameter on the Fermi surface $\delta \Phi(x)$. The mechanism for forming the anomalous Meissner currents is the splitting, $\delta$, of the zero-energy bound states caused either by a finite superflow with momentum, $\mathbf{p}_f = \left(\frac{1}{2} \nabla \phi - \frac{2}{\hbar} \mathbf{A}\right)$, or spontaneous time-reversal breaking by the surface order parameter, or both $\delta \Phi(x), \delta$. The surface currents can be calculated in the Fermi-liquid model of d-wave superconductivity [5,8,9]. This model covers, in general, correlation effects, bandstructure effects, and disorder such as surface roughness [6,11,12]. We use in our calculations the methods described in Ref. [12]. Typical results for the currents near [100] and [110] surfaces, induced by a very small magnetic field ($H = 40\mu T \ll H_c$), are shown in Fig. 3. This figure presents the dependence of the current density on the distance from the surface for various degrees of surface roughness (defined in [12]), and the two typical temperatures, $T = 1K, 18K$. To estimate from these results the screening factor for the experimental setup we consider a representative random distribution of channels shown in Fig. 4, which channels describe the ion tracks and the disordered region around them. The channels are covered in the $a$-$b$ planes by layers of width $\approx 4\lambda_0$ ($\approx 6 \text{ nm}$) of anomalous surface currents. The remaining bulk superconducting regions carry the regular Meissner current. The thickness of our films is $\lesssim \frac{1}{2}\lambda$. Hence, we replace for our estimate the superfluid momentum, $\mathbf{p}_f$, in the film by its average, and obtain the screening factor by adding up the screening currents in the anomalous and regular regions at constant $\mathbf{p}_f$. Of interest is the normalized ratio for a [100] surface, $S_0(T) = S_{[100]}(T)/S_{[100]}(18K)$, and a [110] surface, $S_1(T) = S_{[110]}(T)/S_{[110]}(18K)$. From the screening factors we obtain the effective penetration depth, $\lambda \propto S^{-1}$, for channels in [110] and [100] direction, i.e. $45^\circ$ and $0^\circ$. Our results are shown in Fig. 4. The calculated difference, $S_1^{-1}(1K) - S_0^{-1}(1K)$, is $\approx 2\%$ for channels with rough
as an effect of bulk pairbreaking by defects and natural grain boundaries. This effect is not included in our model calculations and reduces the linear T-dependence to a weak T²-dependence. The large surface roughness needed to fit the data is reasonable in view of the transition region of \( \approx 1 - 2 \) nm covering the amorphous channels. We note that roughness and inelastic broadening tends to suppress the surface phase transition reported in \([10]\), which might be the reason why this transition has not been seen in our experiment.

Finally, we note that a close examination of Fig. 2 shows that even the non-irradiated, “as grown”, YBCO films exhibit a weak, but significant, anomaly in the penetration depth. This is true for all the films (more than 50) we have studied so far. Measurements on the same samples by an independent microwave resonator technique at 18 GHz yield identical results and confirm previous findings \([23]\). Films contain natural interfaces, e.g., grain boundaries, which can be expected to give rise to anomalous Meissner currents analogous to the anomalous currents reported here for irradiated samples. This may explain why the linear temperature dependence of the penetration depth predicted for ideal d-wave superconductors, and reported for pure YBCO single crystals \([21]\), is only seen in thin films of very high quality \([22]\).

We interpret the good qualitative agreement of our systematic measurements of the size and orientation dependence of the anomalous screening effect with the theory of Andreev bound states in d-wave superconductors as an indication of the presence of current carrying Andreev bound states at surfaces of YBCO.

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17 B. Kubis, FZ Jülich, IFF, unpublished.