Observation of symmetric patterns in the flux distribution of thin $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films

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We have investigated the magnetic field distribution in epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films on square SrTiO_3 and MgO substrates. In fields between 20 mT and 1 T a magneto-optic method was applied, complemented by a scanning micro Hall probe for the low field range. The flux is observed to penetrate in a rather regular way, displaying the fourfold symmetry of our samples. The penetration starts preferentially from the sample edges. A qualitative description of this penetration process is suggested, which relies on the curvature of the shielding current paths.

1. Introduction

Among the potential applications of high- T_c superconductors thin epitaxial films of these materials will certainly play a prominent role. For this purpose it is essential to have methods available which allow one to check the sample quality on a large scale, simultaneously being fast and contactless. Here we report on measurements with a magneto-optical technique which fulfills these requirements.

We have used this method to study the penetration of magnetic flux and the resulting distribution in square $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films in fields up to 1 T. The measurements show that – in contrast to what one might have expected – the flux starts to penetrate not at the sample corners, but rather in the middle of the edges of the square. Recently there have been several reports about similar investigations [1, 2], although these studies were mainly performed on a smaller spatial scale, ranging from some $10\text{ }\mu\text{m}$ up to 1–2 mm. Our experiments covered the whole sample area of $10\times 10\text{ mm}^2$. They demonstrate that for a homogeneous film one obtains well defined symmetric flux patterns, which reflect the fourfold symmetry of our samples. Imperfections like scratches or inhomogeneities of the films are readily visible. In the low field range ($\approx 10^{-2}\text{ T}$) the magneto-optical experiments were complemented by measurements with a scan-

ning micro Hall probe, which confirmed these results.

2. Experiment

The epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films investigated here were laser ablated onto $10\times 10\text{ mm}^2$ (001) SrTiO_3 substrates [3] or evaporated on (100) MgO . The films on SrTiO_3 had a thickness of about 300 nm, those on MgO 100 nm and 150 nm, respectively. The critical current density of all films was about 10^6 A/cm^2 at 77 K.

The measurements were performed in an optical toploading cryostat down to temperatures of 2 K. Images of the flux distribution were obtained by means of the Faraday rotation of a thin EuS film [4,5]. In contrast to the experiment of Forkl et al. [2] the EuS (without addition of EuF) was not evaporated directly onto the sample, but instead onto a glass substrate, and was subsequently covered with an aluminum film in order to increase the reflectance for the light to be analyzed [6]. This leaves the surface of the superconductor in its original state, an important requirement for future characterization purposes.

Since the magneto-optic experiments were carried out with a HeNe laser, the thickness of the EuS film was optimized with respect to this light source by a

method first introduced by Kirchner [4]. Very good contrast is obtained if the thickness of the EuS film is $\frac{5}{4}\lambda$, where λ is the wavelength of a HeNe laser in the EuS film.

The glass with the sandwich of EuS and aluminum was directly placed on top of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film. In all experiments the superconducting sample and the glass with the EuS film were immersed in superfluid helium. The EuS film was magneto-optically active up to a temperature of $T \approx 12$ K, somewhat lower than the ferromagnetic Curie temperature of 16.9 K reported for this material [4]. This may be due to an excess of Eu, as measured by EDS (energy dispersive X-ray spectrometry), and also to impurity atoms. To check the possible existence of ferromagnetic domains [7,8] we performed a magneto-optic experiment, as described above, without a superconductor in external fields up to 1 T. No structures on a submillimeter range, indicating the existence of a domain pattern in the EuS film, were observed both with the external field applied as well as after reducing it to zero. Therefore we exclude the influence of ferromagnetic domains on the observed magnetic field distributions in the superconductor, at least in the range of our resolution.

The sensitivity of the magneto-optic set-up used here was limited to magnetic fields ≥ 20 mT. Therefore we investigated the low field range below 20 mT in a complementary experiment. A scanning micro Hall probe at a spatial resolution of about 0.04 mm^2 allowed one to obtain field distributions across the whole sample surface by performing xy -scans. Although much coarser than the magneto-optic technique, this method provided a useful extension and verification of the optical results.

3. Results

Figure 1(a–c) shows a set of pictures taken during the increase of the external field up to 0.25 T after zero field cooling (ZFC). The magnetic field was applied in a direction perpendicular to the sample surface. Bright areas in these pictures correspond to regions of high magnetic field. The spatial resolution in the present experiment was $50 \text{ }\mu\text{m}$, a value which apparently can be further improved by proper imaging optics.

In fig. 1(a) the flux expulsion leads to an increase in the brightness of the sample edges. It is important to note that the magnetic field is maximal around the centers of the edges rather than in the corners. On a rough scale, the flux penetrates the film in a uniform way from all four sides simultaneously establishing overall concave flux fronts which display structures in the submillimeter range. The concave flux penetration from the edges of the sample yields a well-shielded region along the diagonals, giving rise to a roughly cross-like pattern of the same fourfold symmetry as the sample (fig. 1(b)). The width of this pattern shrinks as the field is increased, until at 0.25 T it is only a few tenths of a mm wide (fig. 1(c)). At higher fields shielding in this region gradually vanishes, until above 0.5 T we could not detect any influence of the superconductor in our pictures. These fields also lead to a saturation in the remanent flux distribution, which remains unchanged even after applying fields as high as 1 T.

The remanent flux distribution after applying 0.25 T is shown in fig. 1(d). The largest amount of trapped flux is observed around the region which formerly displayed the best shielding.

To make sure that all the features described above correspond to the superconducting sample (and not to the EuS film) we have performed a second run with the sample being rotated by an angle of 90° . The results were identical to the previous ones, but the images were rotated in the expected manner.

In fig. 2 we show for comparison results for a second sample which appears to be less homogeneous. Figure 2(a–c) again presents the increase of the field from zero to 0.2 T after ZFC. As before the flux fronts are found to penetrate the sample from the edges, and the best shielded regions are again roughly along the diagonals of the sample. Apparently the flux penetrates preferentially from the right side, and has a more inhomogeneous distribution compared to sample 1. The inhomogeneities lead to a distortion of the shielded cross-shaped structure. The remanent flux distribution after reducing the field to zero is shown in fig. 2(d). Again the largest amount of flux is trapped around the regions which were formerly the best shielded ones.

Results for the second sample in the low field region, obtained with the Hall probe, are presented in fig. 3. The sample was again cooled in zero field, and

Fig. 1. Flux distribution in an epitaxial 300 nm thick $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film on a $10 \times 10 \text{ mm}^2$ (001) SrTiO_3 substrate (sample 1). The sample was cooled in zero field and the pictures were taken during increase of the field up to $B = 0.25 \text{ T}$ (which lasted 2 min). The light intensity distribution corresponds to the magnetic field distribution. The contour of the square film is outlined by the expelled flux, best visible in fig. 1 (a). The applied field was (a) $B = 0.06 \text{ T}$, (b) 0.12 T , (c) 0.25 T , (d) remanent magnetization, $B = 0$.

then exposed to an external field of 1 mT and 19 mT respectively. The curves represent lines of constant field at a height of 0.3 mm above the sample surface, plotted at intervals of 0.1 mT in fig. 3(a) and 1 mT in (b). The location of the sample is indicated by the square. It should be noted that the field values near the centers of the edges exceed the externally applied field by about 10% whereas the field above the corners of the sample is nearly identical to the external field. The inhomogeneity of the sample pointed out already in fig. 2 is hardly discernible at 1 mT but leads to quite a pronounced distortion in a field of 19 mT at the right side of fig. 3(b).

4. Discussion

The observed overall patterns are obviously a geometric effect, to be explained by the square symmetry of our samples. The pictures are reminiscent of a magnetooptic experiment by Huebener et al. [6], where rectangular Nb-foils and -films displayed similar structures. There the structure was attributed to domains with different direction of magnetization, comparable to ferromagnets.

For the structures observed here in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films this explanation can be ruled out: if there were domains the magnetization in neighboring areas would have to be of opposite polarity. This was not the case, as could be shown by determining the sign

Fig. 2. Flux distribution in an epitaxial 300 nm thick $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film on a $10 \times 10 \text{ mm}^2$ (001) SrTiO_3 substrate (sample 2). The sample was cooled in zero field and the pictures were taken during increase of the field up to $B=0.2 \text{ T}$. (a) $B=0.04 \text{ T}$, (b) 0.12 T , (c) 0.2 T . (d) remanent magnetization, $B=0$.

of the magnetization with magnetooptics (turning the analyzer). In addition the measurements with the Hall probe clearly revealed that the sign is the same in all four quadrants, both for shielding and for the remanent magnetization.

In our interpretation the local variations in the field distribution resulting from the flux expulsion are responsible for the occurrence of the cross-like structures. As was noted in the discussion of fig. 1(a) and the xy -scan with the Hall probe (fig. 3), the field appears to be maximal around the centers of the sample edges. Therefore this is the location where the lower critical field is reached first, giving rise to the observed concave flux fronts penetrating the sample. Consequently the shielding current path is deformed

as sketched schematically in fig. 4, where in addition the field generated by the shielding currents is represented by small circles. The shielding efficiency of the currents is obviously dependent on the sign as well as on the magnitude of their curvature. As suggested by fig. 4 the shielding of the corners is better than in the middle of the edges. Moreover it is seen that on the outside of the current path, where the field of the shielding current *adds* to the external field, the largest enhancement is expected near the middle of the edges, as observed experimentally. The persistence of sharp structures along the sample diagonals, pronouncedly visible in figs. 1(c) and 2(c), can in this picture be explained by the strong magnetic repulsion of two current paths with opposite direction.

Fig. 3. Field distribution of sample 2 in the low field range obtained with a scanning micro Hall probe. The curves indicate lines of constant field at a height of 0.3 mm above the sample surface. (a) $B = 1$ mT, (b) $B = 19$ mT. The numbers refer to the difference between the local magnetic field and the externally applied field (in mT).

Fig. 4. Schematic sketch of the current path in an intermediate magnetic field where the flux has already started to penetrate the sample. The small circles represent the magnetic field that is generated by the current.

A more quantitative description taking into account the different components of $\text{rot } \mathbf{B} = \mu_0 \mathbf{j}$, as was reported by Baczewski et al. [9], is in preparation.

Inhomogeneities in the sample give rise to a deviation of the shielding structure from a perfect cross. We have investigated several samples with a varying degree of homogeneity in the superconducting quality. The according degree of distortion of the cross was clearly related to the overall sample homoge-

neity, not taking into account structures on a sub-millimeter scale. The appearance of the pattern does not depend on the substrate, nor on the preparation process, as can be seen in fig. 5 for a thermally evaporated film on MgO. Altogether one can thus take the deviation of the magneto-optic pattern from the fourfold sample symmetry as a measure of the overall sample inhomogeneity.

Fig. 5. Flux distribution in an epitaxial 100 nm thick $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ film on a $10 \times 10 \text{ mm}^2$ (100) MgO substrate, after zero field cooling. The externally applied field is 60 mT. The low contrast of this picture is due to the rather homogeneous flux distribution.

5. Conclusions

In summary we have investigated the flux distributions of epitaxial $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films with a contactless magnetooptic experiment in fields up to 1 T and in addition with a scanning micro Hall probe in the low field range. A strong influence of the sample geometry on the flux penetration process is observed. In contrast to recent experiments by Forkl et al. [2], we have found the flux to penetrate our samples in a rather regular way, yielding nearly symmetric shielded regions, roughly along the sample di-

agonals. In the remanent state the area close to the structure appears to be the region where the flux is pinned most effectively. We attribute the observed way of penetration to the influence of the curvature of the current path on the shielding properties. This method appears to be a sensitive check of the homogeneity of superconducting films.

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