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Equilibrium magnetic properties and Meissner expulsion of magnetic flux in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ single crystals

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Abstract

The unusual spatial transformation of the magnetic-flux structure in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ single crystals during magnetization of zero-field cooled samples and during field cooling (the Meissner flux expulsion) is directly visualized by means of the magneto-optical technique. In both cases similar inhomogeneous flux distributions are observed at high temperatures in a magnetic field applied perpendicularly to the basal plane: The magnetic field in the crystal center is approximately equal to the applied field, but at the crystal perimeter a belt of lower field occurs. A model of such an equilibrium flux structure is developed taking into account the real shape of the crystals. The distribution of the Meissner screening current in this geometry is directly measured for the first time.

1. Introduction

Investigations of the magnetic properties of high- T_c superconductors are very complicated because of their strong anisotropy and crystal inhomogeneity, the great influence of thermal effects, the combination of strong and low pinning in the same material at different temperatures, and sample forms which make the interpretation of magnetic measurements difficult [1,2]. Compared with ordinary integral measure-

ments, the direct observation of the magnetic-flux structure in superconductors by means of the magneto-optical technique [3] provides much more detailed information to obtain an appropriate theoretical model. The magneto-optical technique was largely developed in the past years as it was applied to high- T_c superconductors [4–7]. Besides the effects of vortex pinning, important features of the equilibrium properties of the vortex lattice have been found by means of this method. The Meissner expulsion of the magnetic flux from the samples during field cooling was observed in sintered materials [8] and $\text{YBa}_2\text{Cu}_3\text{O}_7$ (YBCO) single crystals [9–11]. In the latter case an inhomogeneous flux distribution was

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revealed: The flux escapes mainly at the perimeter of the crystals. This effect was qualitatively explained by flux trapping in the central part of the crystal due to pinning which also increases with cooling [12–14]. But this explanation only works correctly when the magnetic field is applied parallel to the basal plane of the crystal. In this case the theoretically predicted characteristic depth of the flux expulsion and the field dependence of the Meissner fraction were directly measured [15,13]. For the perpendicular magnetic field used in magneto-optical experiments, the theory has to be modified so as to take into account the demagnetization factor [9,10]. Estimates from the model [12–14] for an applied field of 100 mT give a depth of the flux escape of the order of 1 μm , but the width of the observed area of the Meissner effect was of the order of 100 μm , i.e. much larger than was expected from theory and only five times smaller than the sample width.

Other recently observed manifestations of the equilibrium properties of the vortex lattice are a sharp magnetic wall moving through the superconductor in low fields [16,11] and steps of the penetrating magnetic field on the sample edges due to a Meissner surface current [17]. Further investigation of these related effects is beyond this work and will be published elsewhere. In the present paper we visualize the magnetization of high-quality $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_x$ (BSCCO) single crystals. These crystals have much lower pinning than YBCO crystals, practically vanishing at temperatures higher than 30 K. This allows us to investigate the equilibrium magnetic properties almost without any interference of pinning effects. Both during field cooling (FC), when flux escapes from the crystal, and during magnetization after zero-field cooling (ZFC) when flux penetrates into it, we observe at high temperatures the belt-like structure as seen in YBCO crystals after field cooling [9,10]. This independence from the direction of flux motion shows that there is another reason for the appearance of the “belt”, namely, the equilibrium magnetization of a flat crystal in a perpendicular field. The pinning in this case plays a secondary role, in contrast to the longitudinal magnetization discussed in refs. [13] and [15]. To explain the equilibrium “belt” structure we propose a model taking into account the *rectangular* cross-section of the crystals. Note that the magnetization of a crystal with a *ellipsoidal* cross-section

should be homogeneous. The Meissner surface currents caused by the difference of the internal magnetic field in the flat superconductor and the applied perpendicular field is the main driving force in the investigated processes of vortex motion. We measure the distribution of this current for the first time directly using the recently developed method [18].

2. Experimental

A description of the modern magneto-optical techniques for visualization of the normal field component B_z can be found in refs. [4–7]. As the magneto-optical indicator we use a special high-sensitive ferromagnetic garnet film with in-plane anisotropy [7]. The garnet indicator film grown on a transparent gadolinium–gallium garnet substrate is directly placed on top of the superconductor (Fig. 1). The declination of the spontaneous magnetization M in the film caused by the normal field component is visualized by the Faraday rotation of polarized light which is reflected from an aluminium layer on the bottom surface of the indicator and transmitted twice through the film. Only such an indicator can be used for our experiment at high temperature. The recently proposed frozen-flux technique of high-resolution observations at high temperatures using EuSe layers [11,16] cannot be applied in the temperature region where the Meissner expulsion occurs. Moreover, the sensitivity of EuSe is not high enough for the present measurements.

The polarizers of the microscope were uncrossed to obtain an approximately linear optical contrast. The lower the local field over the sample, the darker the corresponding image on the presented photographs. The field profiles were measured on the video-images transmitted to a PC from the video-processor Hamamatsu DVS-3000.

To measure directly the current distribution the new compensation magneto-optical technique [18] is used. This technique works as follows. In the garnet indicators there are domains with different orientations of the spontaneous in-plane magnetization M (Fig. 1). If the in-plane field B_x above the superconductor changes sign, a tooth-like boundary forms between domains of opposite M on the line of zero B_x . Such an in-plane field gradient is induced by the

Fig. 1. Scheme of the experimental procedure. A flat superconductor (1) is covered by a magneto-optical garnet indicator (2) with a tooth-like boundary separating domains of opposite magnetization. In the forefront the current distribution j induced by the vertical applied magnetic field B_a is shown. The corresponding field rounding the superconductor is presented in the background of the figure. The normal component of the field B_z tilts the magnetization M in the indicator away from the in-plane easy direction of magnetic anisotropy. The shift x_i marks the equilibrium position of the domain boundary where $B_x = 0$ (dashed-dotted line) after applying an additional horizontal field B^I .

currents that screen the magnetic field applied perpendicular to a flat superconductor strip, as shown in Fig. 1. For a thin sample (thickness $d \ll$ half-width a) the in-plane field B_x reflects to an accuracy of d/a the current density integrated over the sample thickness, $J(x) = \int_{-d/2}^{d/2} j(x, z) dz = 2B_x(x)/\mu_0$. So, by measuring the distribution $B_x(x)$ we obtain the distribution of the sheet current $J(x)$. Such an experiment can be carried out by probing the position x_i of the domain boundary when it moves by the application of an additional in-plane test field B^I . Note that the normal flux distribution $B_x(x)$ does not change. The position x_i is determined by the condition $B^I = B_x(x_i)$ (Fig. 1). Thus, we observe the line where the sheet current has just the value corresponding to $2B^I/\mu_0$. Changing B^I we get the current-density profile across the whole sample width.

In the experiment we use big $\text{Bi}_{2.10}\text{Sr}_{1.89}\text{Ca}_{0.99}\text{Cu}_2\text{O}_x$ single crystals of very high quality ($T_c = 86$ K), with

much higher homogeneity and almost without defects compared with the BSCCO crystals tested before by magneto-optical observations [19–20]. The flat crystal plates have sharp rectangular edges like YBCO crystals, but unlike most BSCCO crystals which frequently have rather irregular edges. These sharp edges might be the reason for the clear manifestation of the peculiar flux expulsion in the Meissner state, as will be shown below. All crystals exhibit equivalent properties. For the presentation we choose one of them with dimensions $5.2 \times 1.9 \times 0.086$ mm³. The applied magnetic field B_a was directed along the c -axis and typically ranged up to 200 mT.

3. Results and discussions

Above T_c the external magnetic field magnetizes the indicator homogeneously, and the entire image has a constant brightness. While cooling the BSCCO crystal under the indicator an inhomogeneous structure appears just upon crossing T_c , precisely as was observed on pure YBCO crystals [9,10]. Partial Meissner expulsion of the flux from the crystal is imaged as the darker areas of the indicator corresponding to regions where the magnetic field is lower now (Fig. 2(a)). The contrast of the effect increases with decreasing temperature, stabilizes at about 10 K below T_c and does not change further at lower temperatures. As for YBCO crystals, the highest value of the Meissner fraction $\eta = (B_a - B_z)/B_a$ for the investigated BSCCO crystals is observed near the crystal edges as a dark stripe along the perimeter (Fig. 2(a)). In the central part of the crystals the magnetic field is only slightly lower than B_a . This is clearly seen from the field profile (Fig. 2(b)) determined along the line across the crystal edge indicated by the arrow in Fig. 2(a). B_a is indicated by the dashed line. The shape and amplitude of the profile practically do not change with applied field, so that the Meissner fraction η is lower for the higher B_a . The magnetic field at the edge outside the BSCCO crystal is observed to be enhanced as expected for a flat superconductor expelling the magnetic flux. This positive peak of B_z was not detected in previous investigations on YBCO, possibly due to the fact that the used crystals had a much smaller size in the basal plane and approxi-

Fig. 2. Magnetic structure observed in the BSCCO crystal after field cooling in a perpendicular field of 5.6 mT from above T_c . (a) Micrograph of a part of the crystal at $T = 5$ K. The dark belt at the crystal perimeter is the Meissner flux expulsion revealed by the magneto-optical garnet indicator. (b) The flux profile measured along the line marked by the arrow x in (a). Negative x corresponds to the sample exterior.

mately the same thickness, i.e. a lower demagnetization factor.

The value of the Meissner expulsion depends on the defect structure of the crystal. In a BSCCO crystal from the same family irradiated by heavy ions to produce stronger pinning we do not detect the expulsion. The effect is also absent in dirty YBCO crystals. This is in accordance with the drastic lowering of the Meissner fraction observed in YBCO crystals doped with Ca, Mn and Si, or in twinned regions compared with untwinned regions [10]. So, the enhancement of the pinning force by any kind of defect introduced

into the crystals, reduces the Meissner flux expulsion. But the pure BSCCO crystals investigated at high temperatures are expected to exhibit very low pinning so that its role may be neglected compared with the equilibrium properties of the vortex lattice.

To reveal any irreversibility of the flux behavior we compared the field patterns of the FC experiment with that of the ZFC experiment for the same applied fields. Fig. 3 shows the flux penetration into the same BSCCO crystal of Fig. 2 upon the temperature increase from 5 K to T_c after a constant field of 5.6 mT was applied to the ZFC sample. At low temperatures the applied field is not high enough to magnetize the superconductor, and full screening is observed (Fig. 3(a)). The crystal is imaged as the dark rectangle in which the field is zero surrounded by a field which increases near the crystal edge due to the demagnetization factor.

When the temperature is increased flux enters into the sample in a quite unexpected way. It does not penetrate from the edges squeezing the Meissner phase to the center like is assumed for the Bean model [21] but quite oppositely, the flux appears in the crystal center and moves the Meissner phase to its perimeter (Fig. 3(b)). In the actual situation vortices penetrate through the Meissner phase along defect regions, one of which is seen in the upper-right corner of the crystal, but as will be shown below, such penetration can even take place in an ideal crystal where the Meissner screening currents cause a driving force.

Further increase of the temperature leads to the formation of the same "belt" structure (Fig. 3(c)) as was seen after field cooling (Fig. 2(a)). Qualitatively, the corresponding field profiles (Fig. 3(d)) already obtain the shape of the FC profile (Fig. 2(b)) at $T > 45$ K. But only at $T > 75$ K the amplitude of the inhomogeneity measured as the value of the flux concentration near the edge, $\Delta B = B_z - B_a$, approximately becomes the same as for field cooling (Fig. 3(e)). So, in this high-temperature region, the vortex motion in opposite directions is reversible, and, consequently, the observed flux structure is in equilibrium.

The observed inhomogeneous belt-like flux structure which was *unexpected in equilibrium* cannot be described in frameworks of the usual approximation of the crystal shape by an ellipsoid. It is well known

Fig. 3. Penetration of the magnetic flux into the BSCCO crystal during temperature increase in a field of 5.6 mT applied after zero-field cooling. (a–c) Appearance of the magnetic flux in the crystal center (the same area as shown in Fig. 2(a)). Sequential flux structures with increasing temperatures shown for (a) 5 K, (b) 49 K, (c) 70 K. (d) The corresponding flux profiles measured along the same line as in Fig. 2(a) at temperatures 40, 45, 49, 53, 57, 61, 70 and 80 K. $x=0$ is the crystal edge. (e) Temperature dependence of the flux density near the crystal edge (the height of the peak at $x \leq 0$ in (d)). The open triangles denote $\Delta B = B_z - B_a$ at the peak position during heating of the ZFC crystal in field, the filled circle is the same peak after FC in the same field.

that the equilibrium magnetization of a superconducting ellipsoid is homogeneous and the field inside is constant [22]. The origin of this homogeneous magnetic structure can be explained by the simple scheme shown in Fig. 4(a). The Lorentz force exerted on the flux line ends by the surface screening current $2J_M \times B$ (the factor 2 is due to J_M flowing on both sample surfaces) pushes the vortices to the sample center and just compensates the force from the

vortex lines tension, $T = m_{eq} H_{c1} B$ [23]. Here, like in ref. [13], $\mu_0 m_{eq} H_{c1}$ is the equilibrium magnetization of a type-II superconductor at $H > H_{c1}$, which far above H_{c1} is almost independent of H ($m_{eq} \sim \frac{1}{2}$). The difference between applied field and internal equilibrium field is thus given by

$$\Delta B^{eq} = (1 - N) \mu_0 m_{eq} H_{c1}. \quad (1)$$

Fig. 4. Expected equilibrium magnetic field distributions in a flat type-II superconductor with elliptical (a) and rectangular (b) cross-section with corresponding profiles of the perpendicular field component. The projection of the vortex tension T on the sample surface, which drives the vortex towards the specimen edge against the Lorentz force from the Meissner screening current j_M in the ellipsoid, is absent in the rectangle and is replaced by the driving force due to the gradient of the vortex density.

To screen this field difference one needs a surface current [22]

$$J_M = x \frac{\Delta B^{\text{eq}}}{\mu_0} (a^2 - x^2)^{-1/2}. \quad (2)$$

The inclination of the ellipsoid surfaces $d(x) = d_0 [1 - (x/a)^2]^{1/2}$ leads to an expulsion force

$$T \frac{\partial d}{\partial x} = -m_{\text{eq}} H_{c1} B \frac{x}{(a^2 - x^2)^{1/2}} \left(\frac{d_0}{a} \right), \quad (3)$$

that is precisely the same as $2J_M \times B$ taking into account Eqs. (1) and (2) and

$$(1 - N) = \frac{d_0}{2a}. \quad (4)$$

If the superconductor has a rectangular cross-section as for most high- T_c crystals, there is no inclination of its surface, and, thus, no force from the vortex tension. To maintain equilibrium in this case a flux-density gradient has to appear [24] which drives the

vortices out of the sample against the Lorentz force generated by the screening currents (Fig. 4(b)):

$$d \left(\frac{\partial H}{\partial B} \right)_{\text{eq}} \frac{\partial B}{\partial x} = 2J_M \quad (5)$$

Taking into account that for $H > H_{c1}$ the slope of the equilibrium magnetization curve $\mu_0 (\partial H / \partial B)_{\text{eq}}$ of high- κ materials is approximately 1 and solving this equation together with Eq. (2) we obtain in first approximation for the flux distribution

$$B = B_a - \mu_0 m_{\text{eq}} H_{c1} \left(1 - \sqrt{1 - \left(\frac{x}{a} \right)^2} + (1 - N) \right), \quad (6)$$

where Eqs. (1) and (4) were used. This ellipsoidal flux profile (see Fig. 4(b)) qualitatively coincides with the profiles measured here on BSCCO (Figs. 2(b) and 3(d)) and previously on YBCO [9,10]. Thus, the observed effect can be explained taking only into account the rectangular cross-section of the crystal. Interestingly, at the sample edge we obtain $B - B_a \approx -\mu_0 m_{\text{eq}} H_{c1}$, which is just the equilibrium magnetization of a long cylinder in parallel field. A similar inhomogeneous distribution of the magnetic field was observed in sintered YBCO disks, but in that case the role of equilibrium magnetization $\mu_0 m_{\text{eq}} H_{c1}$ is an average of the individual irreversible magnetizations of single grains [25].

The nonequilibrium increase of the field density at the edges below 75 K compared with the field distribution after field cooling (see Fig. 3(e)) might be due to the effect of the edge barrier discussed by Clem and Huebener et al. [23] in conjunction with the equivalent process of flux-bundle penetration into type-I superconductors [26,27]. Evidence that a barrier for flux entrance plays the main role in the irreversibility of BSCCO crystal magnetization was already obtained by micro-Hall-probe measurements on BSCCO crystals [28,29]. A further magneto-optical investigation of the edge barrier effect in BSCCO will be published elsewhere. We only have to mention here that a more appropriate model for the edge barrier seems to be the model of Provost et al. [30], instead of the model of Clem et al. [23] or the Bean-Livingston surface barrier [31] supposed in refs. [28] and [29]. The edge shape barrier investigated by Provost et al. is not sensitive to imperfections of the

superconductor surface as the Bean–Livingston surface barrier is. It exists only in superconductors with *nonellipsoidal cross-sections* as shown by experiments [30,32]. The appearance of the barrier in the model by Clem et al. [23] calculated for an ellipsoid contradicts these experiments. So the usual rectangular shape of superconducting crystals may play an important role in the irreversibility properties.

In order to complete this picture we give a third illustration of the influence of the sample edge geometry on the magnetization in a perpendicular field. The initial field penetration depth, the initial deviation of the magnetic moment from the ideal screening behavior, and the hysteresis losses calculated analytically in the frameworks of the critical-state model [35] are proportional to the second, third and fourth power of the applied magnetic field, respectively. This result was obtained with the assumption of a constant sample thickness, i.e. again for a rectangular cross-section of the sample. The physical reason is the *additional ability of the sample to carry the critical current in the edge corners*.

The main driving force of the vortex motion in flat superconductors in the perpendicular geometry is produced by the Meissner screening current J_M flowing over the *entire sample surface* (see Eq. (2)). Direct experimental measurements of this nondissipating supercurrent were not available before this work. In refs. [33] and [34] current profiles were indirectly obtained from the measured perpendicular field profiles for a circular superconducting film by solving the appropriate equations. Of course, there is no reason to doubt the validity of such calculations, but it is interesting to demonstrate there for the current distribution under real experimental conditions on a sample of finite sizes in all directions. This is done by means of the new compensation method (Fig. 1) suggested recently in ref. [18] and described above.

The BSCCO crystal was cooled down from above T_c in a perpendicular field of 14.2 mT. The field was switched off at $T=5$ K, where pinning is so strong that the flux is rigidly frozen in, preventing any changes of the flux distribution during observation. After determination of ΔB a separate set of coils produced the additional in-plane test field $B^{\parallel}$ without changing the sample temperature. The position of the magnetic domain boundary in the indicator as a function of $B^{\parallel}$ is presented in Fig. 5. The experimen-

Fig. 5. Distribution of the in-plane field B_x directly representing the Meissner currents screening an applied field of 14.2 mT, measured by the position of the domain boundary in Fig. 1 as a function of the test field $B^{\parallel}$. Open triangles are the experimental points, the curve is calculated from the classical formula (Eq. (2)) for the screening current distribution in an infinitely thin strip.

tal points are in very good agreement with the theoretical curve given by Eq. (2) without using any fit parameters.

4. Conclusions

The origin of the inhomogeneous flux distribution in the equilibrium state of a nonellipsoidal type-II superconductor has been found. This is the central result of this paper. The observed effect is the consequence of an *effectively lower demagnetization factor* N of the additional material near the rectangular sample edge compared to N of the inscribed ellipsoid. Because sufficiently above H_{c1} the magnetization of a type-II superconductor is almost field independent, the field profile should not change with field increase and the Meissner fraction should be $\eta \propto 1/B_a$ (see Eq. (6)). In fact, this had already been observed in ref. [10] with good accuracy.

The visualized inhomogeneity of flux *above the irreversibility line* can explain the reverse asymmetry of the microscopic field distribution of decoupled vortices detected in this region by the muon spin rotation (μ SR) technique [36]. The equilibrium reduction of the field near the rectangular crystal edges (and each surface step also) observed magneto-opti-

cally corresponds to the low-field tail of the μ SR field distribution, whereas the high-field tail of the linear vortex cores disappears above the decoupling (1D melting) line.

The usual rectangle shape of the sample edges producing an edge barrier results in an unusual penetration of the flux into a type-II superconductor with low pinning. The flux directly penetrates to the sample center and a Meissner phase is maintained at its perimeter. This is quite opposite to the way of flux penetration described by the Bean model, which is applicable to superconductors with strong pinning. This unexpected penetration of the vortices through the Meissner phase is due to the Meissner screening current (2) near the sample edges $x = \pm a$, which is directly measured in this work. This current and, thus, the driving force which pushes the vortices towards the sample center appear over the whole sample immediately after application of an external field and strongly diverges at the edges. This inhomogeneous force in a homogeneous superconductor without pinning produces the flux density at its center.

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