# Measurement of the magnetic flux distribution over the surface of a superconducting $YBa_2Cu_3O_y$ single crystal by an ESR method

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The ESR signal from a DPPH grain $\sim 0.06$ mm in size at the center of a platelet of a high-$T_c$ superconducting $YBa_2Cu_3O_y$ single crystal was measured at a frequency of 9.3 GHz. Below the superconducting transition temperature $T_c = 92$ K, the ESR line broadens and shifts. Its width and position depend on the cooling conditions and on the direction of the external magnetic field. The results found can be described well by the critical-state model. This interpretation yields a value $J_c \approx 1.2 \cdot 10^6$ A/cm$^2$ at $T = 4.2$ K. The high sensitivity of this measurement to variations in the magnetic flux made it possible to detect a pronounced irregularity in the distribution of the critical current density in the single crystals.

## 1. INTRODUCTION

Rakvin *et al.*[1] have proposed a new application of the ESR method in research on superconductors. They studied the ESR of $\alpha$, $\alpha'$-diphenyl-$\beta$-picrylhydrazyl (DPPH) deposited on the surface of a superconducting $YBa_2Cu_3O_y$ ceramic. Below the superconducting transition temperature $T_c = 92$ K they observed a line broadening. The linewidth varied from 10 Oe at $T_c = 92$ K to 40 Oe at the lowest temperature attained, $T = 75$ K. This broadening was attributed to a spatial nonuniformity of the magnetic flux distribution in the superconducting mixed state in the magnetic-resonance field $H \approx 3000$ Oe. The magnetic-field penetration depth was found from the temperature dependence of this broadening. An expression for the second moment of the shape of the ESR line, which is determined by the nonuniform distribution in a fluxoid, was used in this procedure.[2]

In our own study, we first attempted to deposit DPPH on the surface of $YBa_2Cu_3O_y$ single crystals consisting of thin platelets grown by the standard procedure.[3] The linewidth of the DPPH crystals was $\approx 1.5$ Oe; it increased to 3 Oe after the preparation of a film from these crystals. As the temperature was lowered in a magnetic field perpendicular to the plane of the platelet, with a strength corresponding to the beginning of the recording of the absorption line, the line initially broadened and split into several components in the superconducting state. There was essentially no change in the position of the center of the line. The linewidth did not tend toward saturation at low temperatures. The latter result contradicts the interpretation of Rakvin *et al.*,[1] according to which the broadening results from a decrease in the penetration depth of the magnetic field in a fluxoid. It is natural to suggest that the broadening actually results from a nonuniformity of the magnetic field produced by the Bean currents.

Our next step was to cement a DPPH grain $\approx 0.06$ mm in size in place of the film at the center of the surface of a single-crystal $YBa_2Cu_3O_y$ platelet, with dimensions of $1 \times 0.55 \times 0.02$ mm. To reduce the spatial nonuniformity of the magnetic field produced by the Bean currents in the direction normal to the surface of the platelet, we fabricated a sandwich of two single crystals bracketing the DPPH grain. This modification of the procedure turned out to have no effect on the measurement results, so a single platelet was used for all subsequent measurements. The changes in the width and position of the line below $T_c = 92$ observed in the process were far greater than in the case of the film. They were furthermore affected by the magnetic history of the sample. Consequently, the order in which the temperature and the magnetic field were varied before the recording of a line was determined by the required final state of the sample. Before each recording, the sample was heated above $T_c$, the magnetic field was imposed, the sample was cooled to the desired temperature, and the line was recorded. We will use "ZFC" to label the cases in which the cooling was carried out in the residual field of the magnet, 20 Oe, while "FC" means the case in which the cooling was carried out in the field at the beginning of the recording, $\approx 3400$ Oe.

The measurements were carried out by means of an ESR spectrometer at a frequency of 9.5 GHz in a flow-through helium cryostat. In this cryostat, the samples could be heated and cooled from 4.2 K to 100 K in a few minutes. The samples were at the center of a rectangular cavity, at an antinode of the magnetic component of the microwave field. This field was in the plane of the platelet, perpendicular to the static magnetic field in all cases.

When the temperature was lowered in the superconducting state, a pronounced increase in noise was observed in the superconducting samples, particularly the ceramic ones. In addition, a hysteresis appeared in the nonresonant absorption. These factors prevented full exploitation of the sensitivity of the ESR method. We did manage to carry out measurements down to $T = 3.3$ K in our case, despite the small dimensions of the DPPH sample, since the noise increase was negligible.

## 2. EXPERIMENTAL RESULTS

### 2.1. Study of the line position

The resonant field $H_0$ in the normal state ($T > T_c = 92$ K) was 3396 Oe, and the linewidth $\Delta H$ was 1.5 Oe. When the samples were cooled to a temperature $T < T_c$ in the residual field of the magnet, and the field was then swept to record the line, we observed changes in the position and width of the ESR line. The crosses in Fig. 1 show the line positions for various angles and for a final cooling temperature $T = 55$ K. Shown in the same figure, by the solid line, is the function $H(\theta) = H(\theta = 90°) + [H(\theta = 0°) - H(\theta = 90°)]\cos\theta$, where $\theta$ is the angle between the normal to the surface of the

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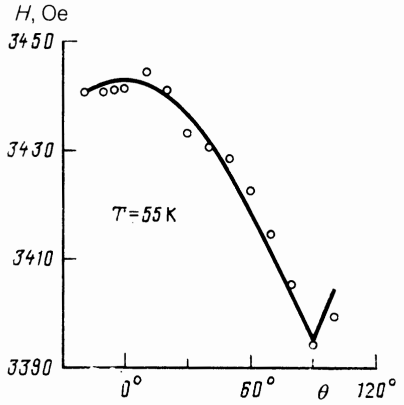


FIG. 1. Orientation dependence of the position of the ESR line found during cooling in the residual field of the magnet, $\approx 20$ Oe, to $T = 55$ K (ZFC).

platelet and the direction of the static magnetic field. We see that the maximum line shift occurs in the direction perpendicular to the plane of the sample and amounts to $\approx 50$ Oe at $T = 55$ K. The "width" of the line, in this case the distance between the outermost extrema of the derivative of the absorption curve, also varied from 3 to 20 Oe. The shape of the orientation dependence $H(\theta)$ and the sign of the shift in the resonant field indicate that the shift of the line is caused by the field produced by screening currents flowing in the plane of the single-crystal platelet.

This assertion agrees with the results of some further experiments in which we studied the temperature dependence of the line shift in the directions perpendicular to the plane of the platelet ($\theta = 0°$, $H_\perp$) and parallel to it ($\theta = 90°$, $H_\parallel$), under FC and ZFC conditions. The dependence for the case of the perpendicular orientation and ZFC is shown in Fig. 2a. We see from this figure that the line shifts up the magnetic-field scale, since the DPPH grain is inside the loop of screening currents. At $T < 60$ K the line shift $h$ is exponential, $\exp(-\alpha T)$, where $\alpha = 6.6 \cdot 10^{-2}\ \mathrm{K}^{-1}$. At $T = 4.2$ K the maximum value is $h = 700$ Oe. In the parallel orientation, under the same cooling conditions, the line shifts in the opposite direction: toward lower fields (Fig. 2b). The reason is that in this orientation the screening currents create a uniform magnetic field in the direction parallel to the surface of the single-crystal platelet. This field is in the same direction as the external field. The DPPH grain in this case is outside

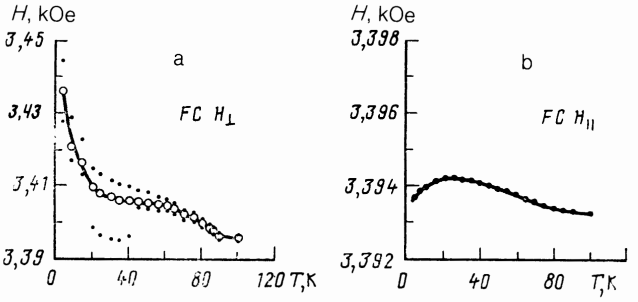


FIG. 3. Temperature dependence of the position of the ESR line found during cooling in the field corresponding to the beginning of the recording, $\approx 3390$ Oe (FC). a—The field is directed perpendicular to the plane of the platelet; b—parallel to this plane.

the loop around which these currents flow. Again in this case we observe a line whose shift with decreasing temperature is comparatively small. The origin of this line will be discussed below. The points bounding the line in this figure indicate the positions of the extrema of the derivative of the absorption line. The distance between these points determines the linewidth. The maximum line broadening observed in the parallel direction was $\approx 10$ Oe.

It is also interesting to examine the behavior of the resonance line in the case in which the changes in the magnetic field during the recording of the line are small, i.e., on the order of the linewidth. The FC cooling regime corresponds to this case. The results of measurements of this sort are shown in Figs. 3a and 3b. We see from Fig. 3b that in the parallel orientation the line shift is on the order of 1 Oe down to the lowest temperatures, and the maximum line broadening is less than 2 Oe. This shift in the perpendicular direction turned out to be unexpectedly large (Fig. 3a), about 40 Oe.

At a temperature on the order of 40 K we see in Figs. 2a, 2b, and 3a some structural features which are associated with either a pronounced broadening of the line (Fig. 3a) or a change in the temperature dependence of the position of this resonance line.

### 2.2. Width and shape of the line

Figure 4 shows some examples of lines recorded during cooling in the residual field of the magnet, for both orientations. In addition to the broadening we see a pronounced diversity in the shape of the absorption curves. Since the

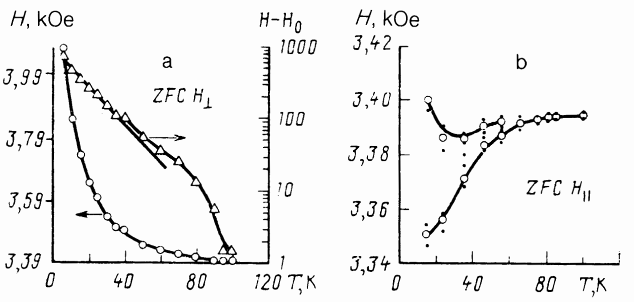


FIG. 2. Temperature dependence of the position of the ESR line found during cooling in an external magnetic field equal to the residual field of the magnet, $\approx 20$ Oe (ZFC). a—The field is directed perpendicular to the plane of the platelet; b—parallel to this plane.

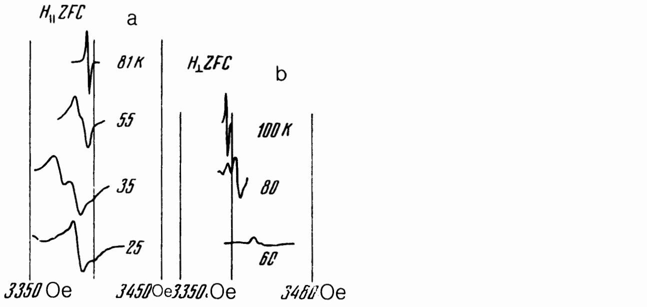


FIG. 4. Representative recordings of the ESR lines during cooling in the residual field of the magnet, $\approx 20$ Oe (ZFC), for the cases in which the magnetic field is (a) parallel to and (b) perpendicular to the plane of the platelet.

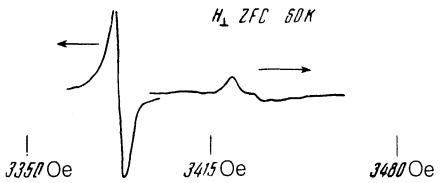


FIG. 5. Demonstration of the sensitivity of the ESR line to the magnetic history of the sample. The high-field line was recorded after the sample was cooled in the residual field of the magnet, $\approx 20$ Oe, and the field was raised during the recording of the line, to 3500 Oe. The low-field line was recorded during the subsequent decrease of the magnetic field.

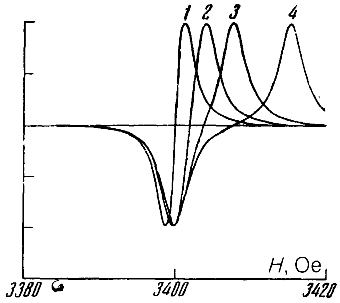


FIG. 6. Calculated derivative of the absorption curves for various magnetic field gradients $k$. 1—$k = 0$; 2—4 Oe; 3—$k = 8$ Oe; 4—$k = 16$ Oe. In the absence of a gradient ($k = 0$), the resonance field is $H_0 = 3400$ Oe, and the line width is $\Delta H = 2$ Oe.

shape of the resonance line is determined by the flux distribution in a region whose size is on the order of that of the DPPH grain ($\sim 0.06$ mm), this diversity indicates a pronounced variation in the flux gradients. To a large extent, these gradients are determined by the uniformity of the distribution of screening currents. Figure 5 shows an example of the sensitivity of the lineshape and of the intensity of the line to the current distribution. The high-field line was recorded in the ZFC regime. During the recording, the magnetic field was raised to 3500 Oe. When the magnetic field was subsequently reduced, the line was recorded again. As expected, it shifted down the field scale. The pronounced narrowing of the line and thus the increase in the line intensity, on the other hand, were unexpected. The explanation of these results may lie in the suggestion of a nonuniform distribution of the critical current flowing near the DPPH grain. During the recording in the reverse direction, this distribution might have become more nearly uniform. The irregular change in the shape and intensity of the line was observed repeatedly, confirming the suggestion that the critical current density not uniformly is distributed in the superconducting single crystal.

To analyze the recordings, we calculated the shape of the ESR line, taking account of the nonuniformity of the external magnetic field. Near resonance, the intensity of the derivative of the absorption line can be described by

$$\frac{dI}{dH} = \frac{2\Delta H (H - H_0)}{[(H - H_0)^2 + \Delta H^2]^2},$$

where $H$ is the external magnetic field, $H_0$ is the resonant field, and $\Delta H$ is the linewidth. The distance between the extrema of the absorption curve is determined by the value $2\Delta H / 1.73$. For the case in which the DPPH is at the center of a uniform circular screening current, the magnetic field distribution in it, $H_{in}(r)$, can be written $H_{in}(r) = H - kr$, where $r = 2R/\xi$, $R$ is the distance from the edge of the sample, $\xi$ is the size of the DPPH, and $k$ is the reduced magnetic field gradient, which is equal to the change in the magnetic field at the position of the DPPH and which is proportional to the critical current. Replacing $H$ by $H_{in}$ in this formula, and summing over the entire volume of the sample (with the help of a computer), we find the absorption intensity as a function of the strength of the external magnetic field. The results of these calculations are shown in Fig. 6. These curves were recorded for $\Delta H = 2$ Oe, $H_0 = 3400$ Oe, and $k = 0$, 2, 8, and 16 Oe. Clearly, the width of the absorption line increases with increasing gradient, and the distance between the extrema is equal to the change in the magnetic field at the position of the DPPH. The low-field maximum tends toward the value of $H_0$, and the position of the line (the zero of the derivative) shifts up the magnetic-field scale, by an amount roughly equal to half the gradient. A gradient of the other sign shifts the line toward a lower field, and the shape of the line is the same as in the preceding case.

### 3. DISCUSSION OF RESULTS

The results of these measurements show that the observed shift and broadening of the ESR line result from the magnetic field produced by the Bean screening current. The calculations of the lineshape confirm that in the case of a uniform current or magnetic-field gradient the position of one of the extrema of the derivative of the absorption line determines the strength of the magnetic field at the edge of the sample, while the distance between extrema determines the nonuniformity of the magnetic field in the sample.

In the case of the parallel orientation, the magnetic field produced by the Bean currents at the surface of the platelet is uniform, and the broadening of the ESR line is caused exclusively by the finite height of the sample above the surface of the platelet. It can be seen from Fig. 2b that the maximum line broadening is less than 10 Oe in this case and is smaller than the broadening observed in the perpendicular orientation. Because of the latter circumstance, we can use the results on the shift of the ESR line (Fig. 2a) to estimate the critical current $J_{c0}$ (A/cm$^2$). The field produced by the current $I$ inside a planar loop of radius $R$ is $h = 2\pi I/(10R) \approx \pi J_{c0} d \cdot \ln(L/2\xi)/5$, where $d$ and $L$ are respectively the thickness and width of the superconducting platelet (both are expressed in centimeters; $J_{c0}$ is expressed in amperes per square centimeter). Substituting in the values $d = 2 \cdot 10^{-3}$ cm, $L = 0.1$ cm, and the shift $h = 700$ Oe found at $T = 4.2$ K, we find $J_{c0} \approx 1.2 \cdot 10^6$ A/cm$^2$. This result agrees with measurements of the magnetization (Ref. 4, for example). We see from Fig. 2a that the temperature dependence of $J_c$ at $T < 40$ K is determined by the argument $\alpha = 6.6 \cdot 10^{-2}$ K$^{-1}$ of the exponential function. This result also corresponds to the value $\alpha = 5.4 \cdot 10^{-2}$ K$^{-1}$ found from measurements of the magnetization of single-crystal platelets.[4]

An estimate of $J_{c0}$ could also be found by working from the measurements of the line broadening. However, we were unable to find the $\Delta H(T)$ dependence, since the line very frequently broke up into several components, or we were able to record only the low-field extremum of this line. Nevertheless, if we calculate the change in the magnetic flux at the position of the DPPH and thus the line broadening

$\Delta H = h\xi/(2L)$, we find $\Delta H \approx 20$ Oe at $T = 4.2$ K. This value corresponds to the maximum line broadening observed in the perpendicular direction.

We regard as the most important result of this study the observation that the ESR line undergoes a splitting, which indicates a nonuniform distribution of the induced current in the superconductor. Experimental support for this interpretation comes from, along with the particular shape of these lines (Figs. 4a and 4b), the existence of an unshifted line in Fig. 2b. This nonuniformity is apparently unrelated to the spatial nonuniformity of the quality of the sample, since it depends on the magnetic history of the sample. The magnitude of the observed nonuniformity is less than the size of the DPPH grain, i.e., $\sim 1$–$10\,\mu$m.

Another unexpected result was the observation of an anomalously large shift ($\approx 40$ Oe) of the ESR line in the case of the perpendicular orientation of the magnetic field for the FC case (Fig. 3a). This result may be evidence that the Meissner state in strong magnetic fields ($\sim 3000$ Oe) is not uniform and that Bean currents are excited in a static magnetic field as the temperature is lowered. Support for this possibility comes from the fact that the magnetization curves $m(T)$, $H =$ const, observed with increasing and decreasing temperature, do not coincide.[5] As was shown in Ref. 6, these currents may be associated with a nonuniform spatial distribution of vortices at the boundary of the sample. In our case, however, this nonuniformity exists at the very center of the platelet.

The last points we would like to call attention to are the abrupt change in the width of the ESR line at $T = 40$ K (Fig. 3a) and the subsequent change in the temperature dependence $H(T)$ (Figs. 2a, 2b, and 3a). This temperature has been mentioned in several previous studies of the temperature dependence of the magnetization.[7]

The new method proposed here makes it possible to study the magnetic flux distribution over the surface of a superconductor in magnetic fields $\sim 3000$ Oe caused by induced superconducting currents. This method has the advantage of a high sensitivity to irregularities in the flux distribution, with the result that one can determine both the critical current and the spatial nonuniformity of the superconducting current flowing near the DPPH indicator sample.

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