

Observation of microscopic currents in superconducting ceramics

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The microscopic screening currents circulating in the grains of the ceramic high- T_c superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ in a magnetic field are visualized by computing the current density with high spatial resolution of more than 10^6 pixels from magneto-optic images taken at the specimen surface with the same high resolution. Having solved several technical and mathematical problems, we can visualize the supercurrent density distribution inside and between the grains of a macroscopic polycrystalline specimen and make quantitative statements. Our method may thus be used to optimize the current-carrying capability of superconducting ceramics. [S0163-1829(98)09817-8]

The discovery¹ of oxides that remain superconducting up to temperatures above the boiling point of nitrogen (77 K) one decade ago caused a boom of research on these high- T_c superconductors. But soon it was realized that one encounters difficulties in using these new ceramic materials, e.g., $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO),² as high-current conductors or in coils for strong magnetic fields, because the loss-free current is limited by grain boundaries and magnetic fields.³⁻⁵ Due to the granularity of these ceramics, the current ceases to be loss free when a relatively low critical current density j_c is exceeded. The loss-free transport current is limited by the Josephson effect⁶ at grain boundaries, where even moderate magnetic fields can suppress the superconducting coupling between the grains. The current carrying capability of a grain boundary is, therefore, an important topic of actual research.^{7,8}

A valuable tool for this investigation is the observation of the penetration of magnetic flux into polycrystalline samples. The penetration of an applied magnetic field can be described as a three-step process.⁹ At low external fields, surface currents screen the sample completely; with increasing applied field, magnetic flux can penetrate the sample along the grain boundaries; and in the third step the flux penetrates into the grains. The macroscopic penetration of flux into polycrystalline YBCO samples was observed previously^{9,10} by magneto-optics, but the corresponding microscopic supercurrents between and inside the grains could not be determined so far.

In this paper we present the visualization of microscopic currents flowing inside and between grains with diameters of 5–30 μm in a YBCO polycrystal. This achievement became possible by recent progress in magneto-optic resolution and by our development of a very effective numerical method which inverts the Biot-Savart law on a grid of over one million pixels to obtain the pattern of currents from the measured magnetic-field pattern. Though the mathematical principles of our method have been known for several years,¹¹ up to now it was not possible to examine complex structures like macroscopic polycrystalline samples quantitatively. We have achieved this now by several improvements, namely, precise gauge of the light intensity as a function of $|H_z(x,y)|$, consideration of the finite thickness of the sample, and elimination of problems caused by the periodic continu-

ation of the measured data. The accuracy and uniqueness of the improved inversion scheme is demonstrated by Jooss *et al.*¹²

The magnetic-flux distribution was measured $\approx 5 \mu\text{m}$ above the sample by a magneto-optical technique.⁹ As a field-sensing layer we use an FeGdY-garnet film¹³ that exhibits a strong Faraday effect. This indicator is placed onto the polished flat surface of the sample. The images are obtained by a polarization light microscope and a charge-coupled-device camera with over 10^6 pixels. The magnetic field was applied after zero-field cooling to 5 K.

The measured perpendicular component of the magnetic field H_z in the plane of our field-sensing film is generated by a current density \mathbf{j} , which is approximately planar. Since our sample is isolated, one has $\text{div } \mathbf{j} = 0$; we may thus describe the current distribution by a scalar potential $g(x,y)$ yielding the two components $j_x(x,y) = \partial g / \partial y$ and $j_y(x,y) = -\partial g / \partial x$. The Biot-Savart law then reads explicitly,

$$H_z(x,y,z) = \int_V g(x',y') \frac{2(z-z')^2 - (x-x')^2 - (y-y')^2}{4\pi|\mathbf{r}-\mathbf{r}'|^5} d^3r'.$$

To obtain the local current density from the measured magnetic-field distribution, we have to invert this equation. This is a large-scale problem due to the large number of $N = 10^6$ datapoints. The standard numerical method of inverting a matrix¹⁴ requires a computation time that grows as N^3 and is thus not feasible even on the largest existing computer (here the matrix has over 10^{12} elements); for an advanced inversion of this particular matrix see Wijngaarden *et al.*¹⁵ However, if we continue the magnetic-field pattern periodically in the x,y plane, we may invert the above relation between H_z and \mathbf{j} by means of a fast Fourier transform with much less numerical effort of only $N \ln N$ operations.^{12,16} The periodic continuation does not introduce noticeable artifacts if we choose the basic area somewhat larger than the specimen area. Using a trick we did not lose resolution by this extended periodicity area. The fact that the current distribution in our disk is not exactly planar, does not disturb either: Our method acts like a microscope with finite depth of focus, depicting the plane closest to the indicator film sharpest. The often discussed general ambiguity of this kind of inversion

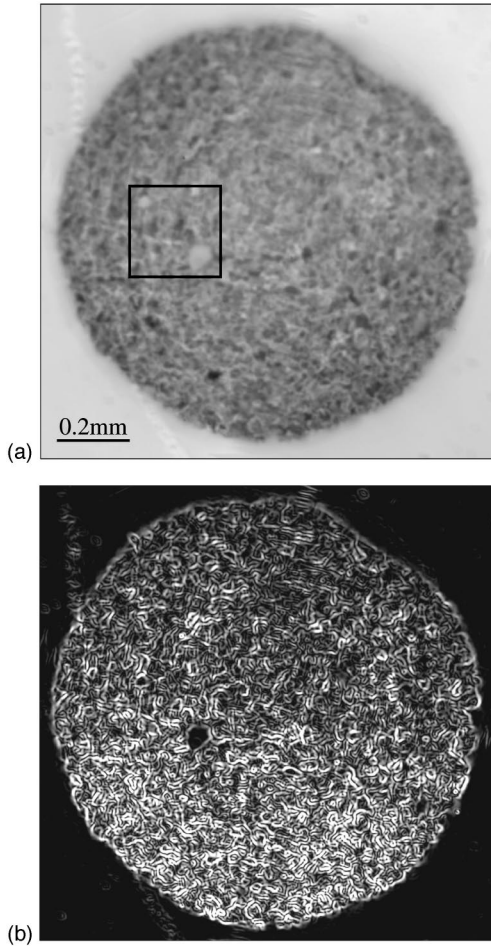


FIG. 1. (a) Magneto-optical image of the flux density distribution of the whole YBCO sample for an external field of $B_{ex} = 64$ mT. The black frame refers to the detail of Figs. 2(a)–2(c). (b) Grayscale image of the absolute value of the flowing supercurrents calculated from (a) by means of the inversion method. The grayscale from black to white corresponds to currents ranging from 5×10^8 to 6×10^9 A/m².

scheme is irrelevant in this essentially two-dimensional situation, as we have checked in detail by performing the reverse Fourier transform for several known current distributions.¹²

For our measurements we use a polycrystalline YBCO sample with diameter of 1.18 mm and thickness of 0.50 mm, prepared by heating a stoichiometric mixture of Y_2O_3 , $BaCO_3$, and CuO in air for 1 h at 870 K and afterwards for 20 h at 1190 K. The powder was then pressed into cylindrical shape under pressure of 8.2×10^5 kPa. After repeating the sintering procedure the sample surface was polished thoroughly.¹⁷

Figure 1(a) shows the measured magnetic-flux distribution for an applied field of 64 mT. To reduce polarization effects caused by the microscope, we subtract the image of the zero-field cooled state. Due to the granular microstructure and the different crystallographic orientations of the grains, the flux distribution is rather inhomogeneous: first the parts of the sample along or near the grain boundaries are filled up with magnetic flux. This is seen as a variation of the grayscale from black to white in Fig. 1(a).

The flux penetration into the sample is directly related to supercurrents. Remarkably, a rather diffuse flux distribution

yields a sharp current distribution. An image of the magnitude $|\mathbf{j}(x,y)|$ calculated from the data of Fig. 1(a) is shown in Fig. 1(b). The grayscale refers to currents ranging from 5×10^8 A/m² (dark gray) to 6×10^9 A/m² (white). This image shows an important achievement of our new technique. From one single image we obtain the entire current distribution in the top layer of a macroscopic polycrystalline sample with spatial resolution of ≈ 5 μ m. All the required information is contained in Fig. 1(a) and no additional assumptions have to be made. Both the intergranular currents at the border of the sample and the microscopic intragranular current loops are observed. In addition to this, precise quantitative statements can be made. In particular, the high spatial resolution of this method allows us to investigate the influence of the microstructure of the material on the current distribution.

In this paper we consider the current distribution near single grains and nonsuperconducting objects, therefore a small section of the sample is depicted in Fig. 2. In Fig. 2(a) the local grain configuration at the surface of the sample is displayed as obtained by a polarization light microscope. Different grayscales refer to different crystallographic orientations of the grains. Figure 2(b) shows the magnetic-flux distribution; bright parts correspond to places totally penetrated by flux, the dark parts are nonpenetrated. In the same image the corresponding current streamlines are depicted. Note that the current streamlines and the contour lines of the flux distribution do not coincide. In Fig. 2(c) a grayscale image of the magnitude of the current density is shown.

In Fig. 2(a) there are three particular objects (A–C) that may be examined further by regarding Figs. 2(b) and 2(c). The nonsuperconducting inclusion (A) and the pore (B) exhibit similar magnetic behavior. Both are penetrated homogeneously by magnetic flux as can be seen in Fig. 2(b). The current density thus vanishes inside the objects A and B. The superconducting environment is screened by intergranular supercurrents with magnitude of $\approx 2 \times 10^9$ A/m², which are clearly visible in Fig. 2(c). However, most of the single grains such as C are not fully penetrated by magnetic flux at the applied field of 64 mT. These grains are recognizable by their dark gray color in Fig. 2(b). The magnetic screening of these grains occurs because of their higher intragranular critical current densities $j_c \approx 6 \times 10^9$ A/m².

When comparing the microstructure and the current flow we realize that by far not all of the grains are surrounded by supercurrents; most currents are flowing in larger loops extending over several grains. This finding is related to the varying capability of different grain boundaries to carry supercurrents: It is known^{18,19} that low-angle grain boundaries can carry critical current densities j_c comparable to those of single grains, while boundaries between grains with large crystalline misorientation exhibit very low j_c . In Fig. 2(d) the microstructure and current stream lines are combined into one picture. The displayed detail is a magnification of grain C and its surroundings. Two kinds of closed current loops are visible: The intragranular current loop inside grain C with high supercurrent density, and current loops extending over several grains and crossing several grain boundaries. Examination of the closed current loop D shows that the current density of intergranular currents is suppressed by a factor of 2 compared with the current density inside grain C. This is shown in the current profile plotted in Fig. 2(d).

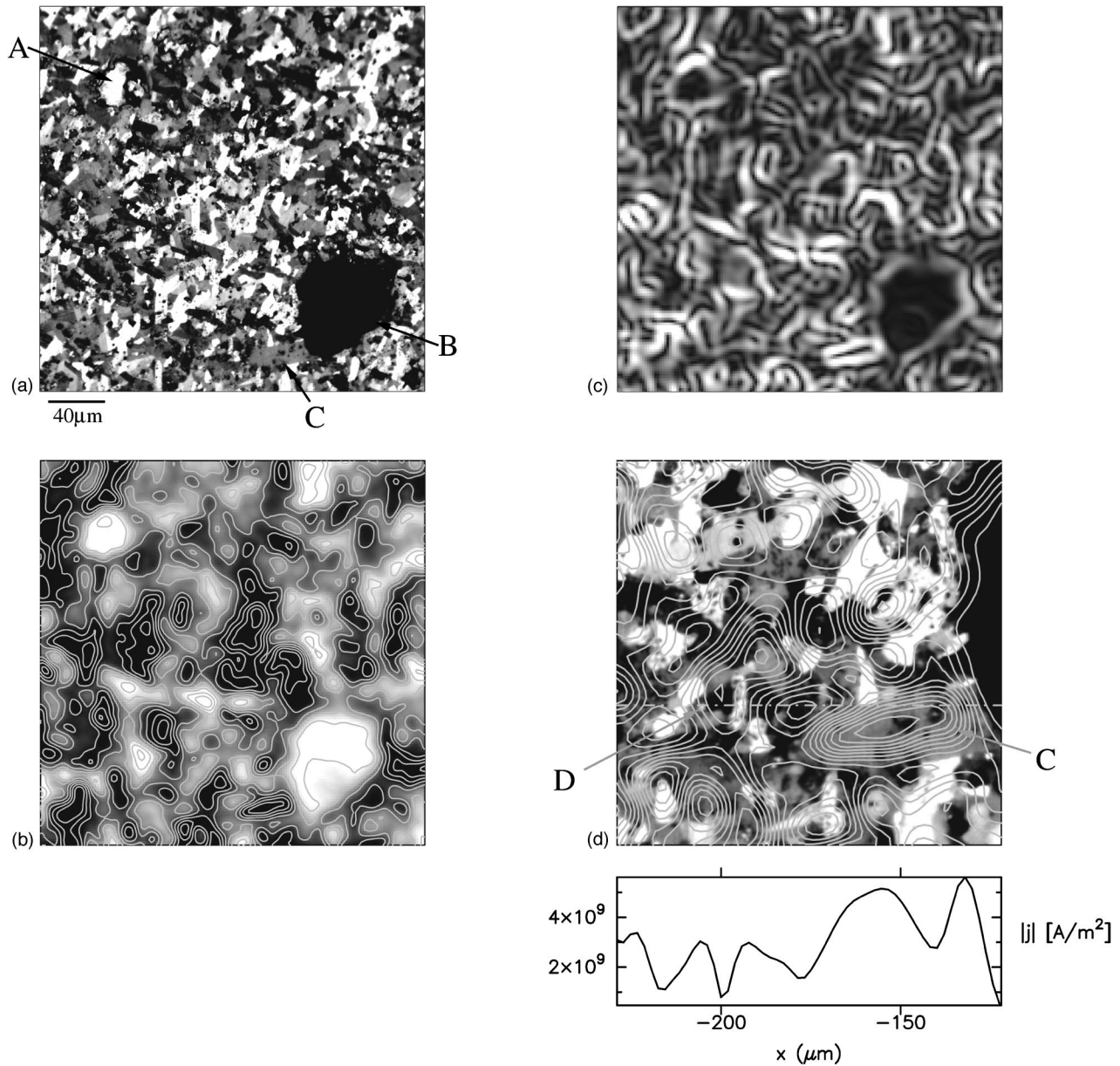


FIG. 2. A detail of the sample shown in Fig. 1(a). Optical image of the grain structure obtained by a polarization light microscope. (b) Flux distribution together with the current streamlines. (c) Magnitude of the currents. The brightest part of the image corresponds to the maximum value of the current density of $5-6 \times 10^9$ A/m². (d) Smaller section showing microstructure and current streamlines. At the bottom we show the profile of the absolute value of the current density taken along the dashed line in the image.

In conclusion, we have visualized the flow of the intragranular and intergranular supercurrents in a polycrystalline superconductor. It is now possible to investigate the behavior of microscopic currents circulating within the grains or crossing grain boundaries. This high-resolution imaging of currents allows us to get a deeper insight into the relation between microstructure and current carrying capability of ce-

ramic superconductors. This should facilitate also a better characterization and optimization of sintering processes during the fabrication of superconductors for practical applications.

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