PHYSICAL REVIEW B VOLUME 36, NUMBER 16 1 DECEMBER 1987

Magnetic microstructure and flux dynamics of high-T_c superconductors

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(Received 21 August 1987)

We present high-resolution Bitter patterns which reveal the magnetic microstructure of the high-T_c superconductor $Ba_2YCu_3O_7$, allowing the direct observation of vortices, and their response to magnetic-field changes. By forming patterns in the presence of a time-varying magnetic field, or a transport current, we also study the dynamics of flux motion. Our results reveal a hierarchy of pinning sites and a remarkable anisotropy in the characteristics of neighboring twins, and allow the mapping of superconducting regions in the presence of circulating and transport currents.

Knowledge of the static and dynamic magnetic microstructure of superconducting materials—the nature and distribution of vortices and their response to magnetic-field changes or transport currents—is of basic as well as technological consequence. In this paper we present high-resolution Bitter patterns[1,2] obtained by the production and deposition of fine (≈ 80 Å diameter) ferromagnetic particles on a superconducting substrate in an applied magnetic field. Under static conditions, we directly image vortices in $Ba_2YCu_3O_7$, and by investigating their response to magnetic field changes, observe a hierarchy of pinning strengths. By forming patterns in the presence of time-varying magnetic fields or transport currents, we study the dynamics of flux motion. The resulting patterns map the flux distribution and motion directly, and, for example, reveal a remarkable anisotropy in the behavior of neighboring twins.

We have used "single-phase"[3] polycrystalline $Ba_2YCu_3O_7$ samples, produced by mixing appropriate powders and calcining at 900 °C, followed by milling, sintering, and annealing treatments. These samples exhibit a sharp (≈ 1 °wide) resistive transition at ≈ 95 K. Optical and transmission electron microscopic (TEM) characterization show them to consist of sintered granules ≈ 10 μm in size, which are composed of ≈ 0.5 μm wide crystallographic grains.[3(b)] The grains in these samples are heavily twinned about (110) planes. On a polished surface, this can be observed optically on a coarse scale, but TEM sometimes reveals microtwinning on a finer scale.[3(b),4] We observe no differences in the internal microstructure of the twins. When examined in a scanning electron microscope (SEM), the twins produce little contrast and are, therefore, barely discernible.

Sample dimensions were $10\times5\times2$ mm^3, except for transport experiments, where the thickness was reduced from 2 to 0.04 mm. Fine Fe particles were produced by evaporation in an Ar atmosphere and deposited on the sample held at liquid-nitrogen temperature. A constant, or time-varying magnetic field was applied perpendicular to the plane of the sample by means of an electromagnet, and a transport current could be passed through the sample by means of surface contacts. Subsequent to particle deposition, the samples were warmed up to room temperature and examined microscopically. All micrographs presented here were obtained in the secondary electron mode of the SEM. It is well established that ferromagnetic particles aggregate primarily on regions of high-flux density, and that particle/substrate interactions are sufficiently strong to prevent rearrangement of the patterns after deposition.[1,2] Evaporation on samples above the critical temperature with or without an applied magnetic field results in a uniform and random deposition of particles. We estimate that the amount of decorating material we use corresponds to a uniform coverage of a few Å. This fact and the results of previous investigations[1,2] make it unlikely that the magnetic microstructure of the sample (with grains ~ 5000 Å in size) is strongly affected by the presence of the decoration. By appropriate choice of field history and deposition conditions, a variety of magnetic microstructural features can be observed. A detailed description of these conditions will be reported elsewhere. Since magnetic decoration experiments delineate the field distribution at the sample surface, the relation of the resulting patterns to the underlying bulk structure must be established in each case. Nevertheless, as we show below, the patterns are informative, and are in some cases directly related to the sample microstructure at the surface. Finally, ferromagnetic particles may be charged and, thus, respond to electrostatic as well as magnetic forces. In the following, we show that our results can be consistently interpreted in terms of magnetic effects alone. But at present we cannot definitively rule out the presence of electrostatic influences.

Figure 1 represents the pattern obtained when the sample is cooled to liquid-nitrogen temperature, a field of 200 G applied and Fe evaporated.[5] The decoration pattern consists of individual bright dots, representing agglomeration of Fe particles, surrounded by a lower deposition of Fe in the form of thin circular sheets, which appear darker than the substrate. (In the secondary electron mode of the SEM, areas decorated with flat layers of Fe appear darker than the substrate, while protruding topographic features, including agglomerations of Fe particles, appear bright. We have confirmed this by control deposition ex-

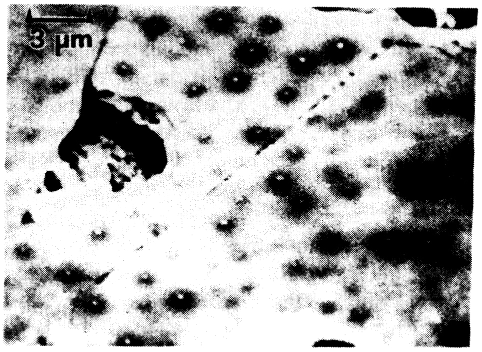

FIG. 1. SEM micrograph showing the decoration pattern in an applied magnetic field of 200 G at liquid-nitrogen temperature. The dots represent agglomeration of Fe particles and are likely to correspond to vortices. The dark lines are boundaries between sintered granules.

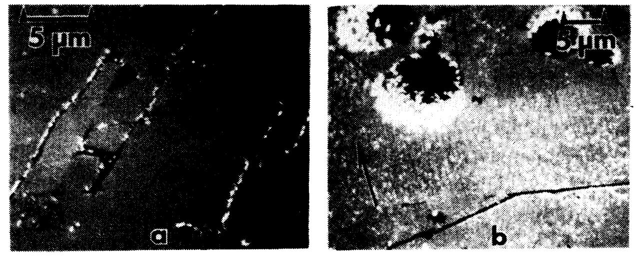

FIG. 2. Decoration pattern after an initial field of 600 G is reduced to 200 G. The presence of bright dots indicates agglomeration of Fe particles protruding from the surface. Note the decoration of some sintered boundaries in (a). In (b) surface cavities are strongly decorated, and in the interior of some grains the decoration is parallel to the trace of the twin planes indicated by the arrow.

periments.) The density of the individual dots is consistent in order of magnitude with the number of vortices expected in an applied field of 200 G in this material, which has a lower critical field of ≈ 150 G at the deposition temperature.[6] We thus believe them to represent the first observation of vortices in high-T_c superconductors. Although a periodic two-dimensional lattice of vortices is in principle expected, it is well known that inhomogeneities can easily prevent the formation of long-range order.[2] The vortex decorations appear to differ somewhat in size. Deposition inhomogeneities, particle-particle interactions, and the presence of more than one unit of flux in some vortices are possible causes.

Increasing the applied magnetic field to a high value and reducing it prior to deposition causes additional flux movement and enables pinning sites to trap vortices. Thus, the strongest pinning sites are decorated first, with increasing deposition leading to decoration of weaker sites. Figure 2 represents the observed pattern when the field is increased to 600 G and reduced to 200 G before evaporation. The strongest decoration often occurs at surface cavities, and sometimes at regions with no topographic features, which may be due to cavities below the surface. Next, boundaries between sintered granules tend to decorate. The grain interiors show the weakest decoration tendency, with the decoration sometimes following the direction of the twins. Thus, cavities, granule boundaries, and twins appear to form a pinning hierarchy of decreasing strength. However, exceptions to this hierarchy are also observed, which may be due to the influence of the underlying bulk material.

The remarkable difference in the flux accommodation properties of the twins can be observed more dramatically in dynamic experiments. Figure 3(a) shows the decoration pattern when the magnetic field is reduced from 200 G at a rate of 5 G/min during the deposition. The pattern reveals the regions traced out by moving vortices and, in effect, produces a long exposure photograph of the moving fluxoids, with the exposure time corresponding to the duration of the deposition (≈ 5 s).[7] Due to the flat nature of the deposited Fe, the decorated regions now appear dark. Twins show strong contrast, but only in regions where the macroscopic field distribution has allowed decoration to occur. Ignoring the small difference in the lengths of the **a** and **b** axes, the twins differ only in the orientation of the basal O-Cu-O chains with respect to the magnetic field.[3(b),8] It is therefore noteworthy that the

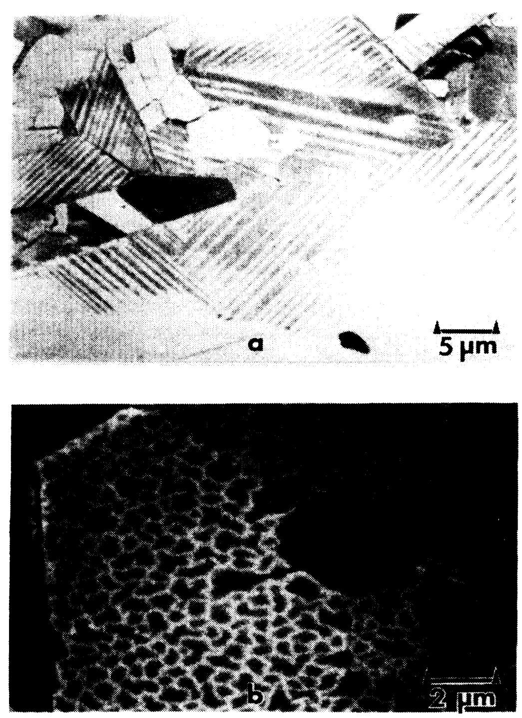

FIG. 3. The decoration pattern formed by evaporation while the magnetic field is being reduced. Decorated regions now consist of flat sheet of Fe and appear dark. They represent the regions swept by the moving vortices during the evaporation. (a) Rate of field change: 5 G/min. Note the strong contrast between the black and white stripes in regions which received Fe particles. The stripes correspond to twins. (b) Rate of field change: 20 G/min.

orientation of these chains plays such a decisive role in the magnetic properties of $Ba_2YCu_3O_7$. These observations also indicate that in this case, the dominant magnetic effect is due to the sample microstructure in the near surface region.

The dynamic behavior of the vortices when subjected to a more rapidly varying magnetic field is shown in Fig. 3(b), where the field is reduced from 200 G at a rate of 20 G/min during a 5 s deposition. The pattern consists of a network of undecorated (bright) regions surrounding dark decorated patches representing the areas swept by the vortices during deposition. The decoration of a substantial portion of the sample is indicative of wide-spread vortex depinning. Although the bright undecorated network of Fig. 3(b) resembles the crystallographic grain boundary structure, our results on several other similar samples show that the networks do not necessarily resemble the crystallographic grain boundary structure observed in the TEM.

The dynamics of flux motion in the presence of a transport current are shown in Fig. 4. A current of 80 mA is passed through a sample $\approx 5\times 0.04$ mm^2 in cross section. This corresponds to a current density of 40 A/cm^2, the nominal critical current density of this material. The pattern consists of dark (i.e., decorated) patches with lines in the approximate direction of current flow. When the current density is increased to 200 A/cm^2, isolated dark patches are the most prominent features [Fig. 5(a)]. This indicates that high-flux gradients only occur over small regions of the sample, where presumably superconductivity is not totally destroyed. The internal structure of some of these decorated patches consists of bright networks, which resemble that of Fig. 3(b), but are elongated in the approximate direction of current flow. The observation of these networks under conditions of both large magnetic field change and transport current, and their elongation with increasing current lend support to the notion that the dark patches represent regions of flux confinement. Thus, vortices are trapped, and under the influence of the changing magnetic field or transport current trace out the dark areas, from which they emerge with difficulty.

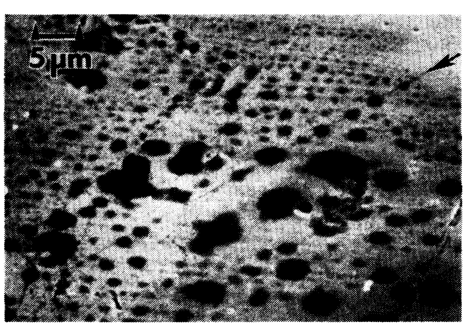

FIG. 4. The decoration pattern obtained when a current density of 40 A/cm^2 is passed through the sample in a field of 200 G. Again the dark regions are decorated and are those swept by the vortices during evaporation. Current flow is from left to right. Note the lines in this direction, and the presence of pinning sites at the arrowed boundary.

Our dynamic observations in the presence of large transport currents provide further evidence for the presence of differences between neighboring twins [Fig. 5(b)]. Since during a dynamic decoration experiment a moving vortex leaves a dark trace behind, the observed SEM contrast between the twins indicates differences in their transport properties. Interestingly, black patches, i.e., strong pinning sites, appear to be present at the sintered boundaries. This suggests that flux motion can begin at a pinning site at a sintered boundary, proceed rapidly along one of the two possible twins, and terminate at another pinning site at the opposite sintered boundary. Thus, Fig. 5(b) offers an intriguing picture of transport in these materials under conditions of technological interest.

In conclusion, we have presented high-resolution Bitter patterns, which directly reveal the static and dynamic magnetic microstructure of the high-T_c superconductor $Ba_2YCu_3O_7$. We have observed vortices and a hierarchy of pinning sites, and investigated the dynamics of flux motion due to changing magnetic fields and transport currents. The results indicate a remarkable anisotropy in the properties of twinned regions and essentially allow the spatial mapping of flux accommodation. Thus, these decoration techniques may also be useful in the spatial localization, and hence identification, of the phases thought to be responsible for the recent indications of superconductivity at very high temperatures.

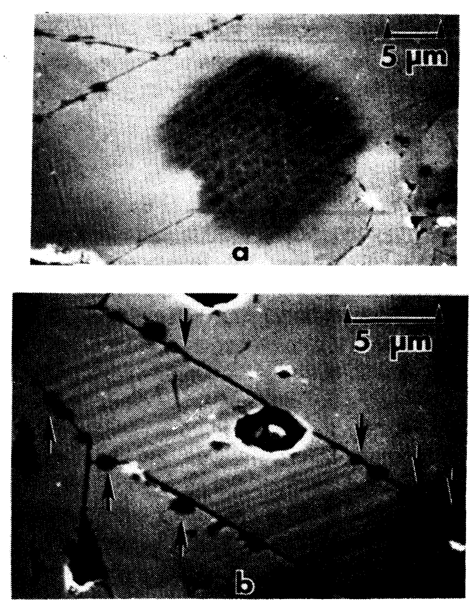

FIG. 5. The decoration pattern formed with a current density of 200 A/cm^2 passing through the sample in a field of 200 G. The decorated areas appear dark. Current flow is from left to right. (a) Isolated, strongly decorated patch. The internal structure of the patch resembles that of Fig. 3(b), but is elongated in the approximate direction of current flow. (b) Vortex motion across a grain. The moving vortices have left decorated dark traces behind, which correspond to one set of twins. Note the presence of black patches, i.e., strong pinning sites (arrowed) at the boundaries.

We acknowledge valuable discussions with A. Ashkin, D. J. Dishop, A. Dayem, R. C. Dynes, G. Fisanick, R. E. Howard, D. W. Murphy, J. C. Phillips, and G. J. Dolan, who also kindly provided a copy of his doctoral thesis.

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[3](a) Although these samples are characterized as single-phase by x-ray diffraction, microstructural analysis shows them to contain compositional inhomogeneities. (b) A. Ourmazd, J. A. Rentschler, J. C. H. Spence, M. O'Keeffe, R. J. Graham, D. W. Johnson, Jr., and W. W. Rhodes, Nature **327**, 308 (1987).

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[5]Due to sample shape effects, the magnetic field at a point on the sample surface may be substantially different from the applied value.

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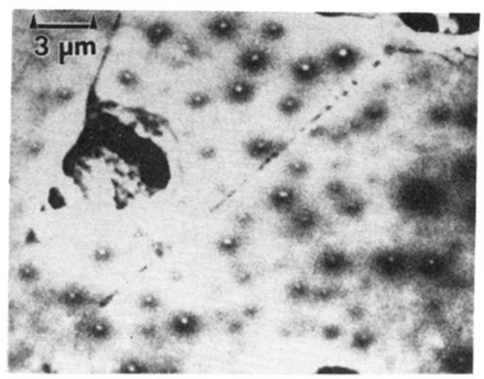

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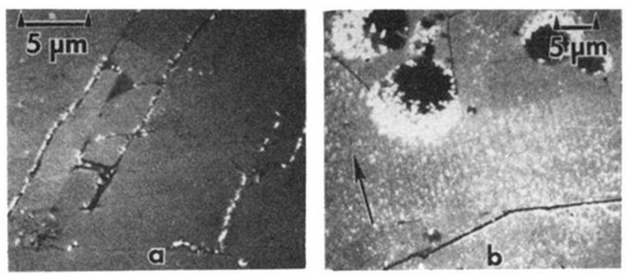

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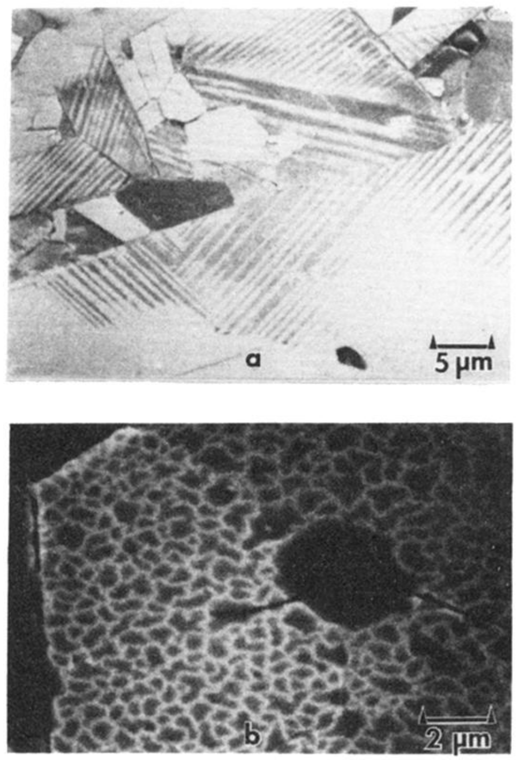

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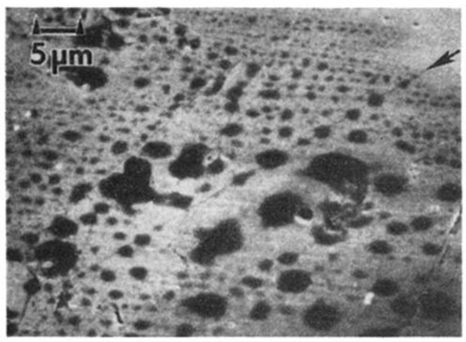

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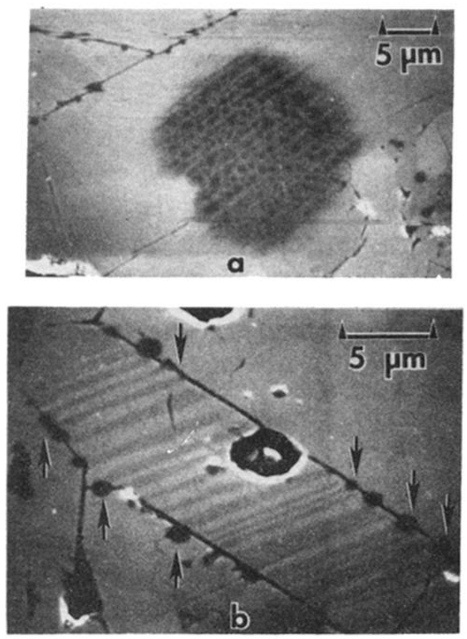

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