

## Flux-Creep Crossover and Relaxation over Surface Barriers in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ Crystals

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Flux creep in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$  crystals exhibits two different regimes as a function of time, as well as of temperature and magnetic field. The short-time, low-temperature regime has a peak in current density versus field, which is enhanced by irradiation defects. The long-time, high-temperature regime has a monotonic and sharp falloff (step) in current density versus magnetic field, which is suppressed by irradiation defects. The former is identified with bulk pinning, and the latter with a surface barrier.

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$\text{BiSrCaCuO-2:2:1:2}$  (BSCCO) crystals are much studied as a model system for almost-two-dimensional superconductivity and for its effects on vortex lattice and flux creep [1–5]. In addition, these materials, in the form of polycrystalline tapes, have great promise for applications [6]. Since flux creep largely controls the current density, there is thus a double interest in the study of flux creep in BSCCO crystals.

While many results have been reported on this system, they form a complex and somewhat confusing picture in which the different regimes are not clearly identified. For example, Zhukov *et al.* [7] and Zavaritsky and Zavaritsky [8] have recently identified two regimes of pinning from the temperature dependence of the current density, but the nature of those regimes is left unclear. Here we present new flux-creep data on what we believe are exceptionally uniform crystals. These data reveal two distinct regimes with qualitatively different behavior. There is strong evidence that the high-temperature, long-time regime is dominated by surface barriers, presumably of the Bean-Livingston type [9–12]. This mechanism has so far been recognized only by Kopylov *et al.* [11] in the BSCCO context, though in detail our results differ significantly from theirs. Our conclusion implies that much earlier theoretical work focusing on bulk mechanisms for current density should be refocused on explaining the low-temperature rather than the high-temperature data. Some aspects of the present work have been presented recently [13,14].

The  $\text{Bi}_{2.2}\text{Sr}_{1.8}\text{CaCu}_2\text{O}_8$  crystals [15] are grown by the floating-zone method and exhibit a sharp superconducting transition at  $T_c \approx 87$  K (varying by 1 K from crystal to crystal). The magnetization, or more properly the stray field proportional to the current density times the sample thickness [16], is measured at the surface of the sample by a miniaturized-Hall-probe technique described elsewhere [17]. This stray field, denoted  $H_s$ , is the difference between the actual value of field recorded on the sample surface and the externally applied field. The sensitivity of this technique permits detailed studies of relaxation in a

low-current-density regime inaccessible in most previous studies. Samples are also irradiated with 2.5-MeV electrons at 20 K, as described earlier [13]. Several BSCCO-2:2:1:2 crystals of area  $1 \text{ mm}^2$  and thickness (along the  $c$  axis) from 20 to  $100 \mu\text{m}$  were studied; results are similar and do not depend in an obvious systematic way on thickness.

The open circles in Fig. 1 show an example of a hysteresis loop measured on an unirradiated crystal at 50 K starting in the virgin zero-field-cooled state, with applied field perpendicular to the crystal face. The approximate time scale for each measurement is about 1 sec. The sharpness of the negative peak in the field-increasing branch at 23 Oe indicates a sample with low bulk pinning and excellent homogeneity. The shape of the ascending-branch data and the flatness of the descending-branch data close to  $H_s = 0$  are evidence of surface barriers, as

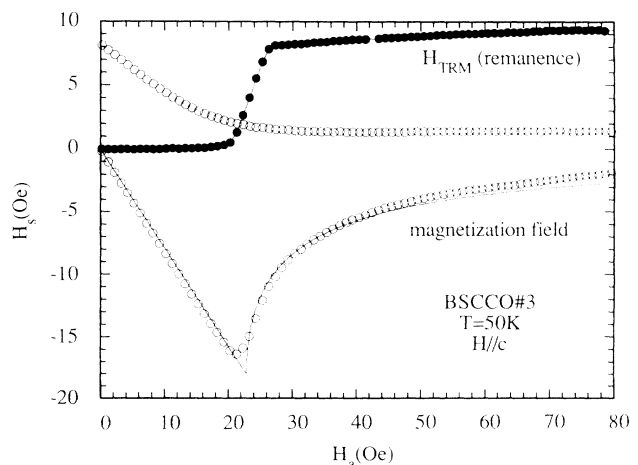


FIG. 1. Surface stray field  $H_s$  of a  $\text{BiSrCaCuO-2:2:1:2}$  crystal vs applied field. Open circles represent a hysteresis loop from the virgin zero-field-cooled state; solid circles represent the remanent stray field (TRM) vs field of maximal excursion of the loop. The solid line above 23 Oe shows a fit by Eq. (1) of the text.

discussed below. The solid circles represent the trapped field, that is, the remanent signal after applied field is reduced to zero, plotted versus the maximum field of the loop excursion. The lack of field trapping below 23 Oe confirms that the linear ascending branch of the hysteresis loop is the Meissner region of full flux expulsion from the sample.

Figure 2 shows hysteresis-loop data on unirradiated and irradiated samples in a somewhat lower-temperature regime, which we call the "crossover regime." Here  $\Delta H_{irr}$  represents the difference between field-increasing and field-decreasing branches, which we interpret as proportional to the in-plane persistent current density. The data present a crossover between a higher-temperature region showing a  $\Delta H_{irr}$  step around 400 Oe and a lower-temperature region showing a peak as a function of field in the same field range. A key discovery of this work is the systematic crossover from peak to step in this temperature range. Furthermore, there is a remarkable contrast in the response of those features to electron irradiation as shown in Fig. 2: The step is suppressed by the irradiation to lower fields and lower  $\Delta H_{irr}$  (lower current densities), while the peak is enhanced to higher  $\Delta H_{irr}$ .

Further insight into this crossover is provided by the flux-creep data in Fig. 3 at two temperatures in this region. The data are taken at constant temperature after dropping this field from about 800 Oe (sufficient to saturate the hysteresis) to the value indicated on the figure. Both sets of data show a crossover as a function of time, with the crossover shifted to shorter times at higher temperature. This is one of the key discoveries of this work, namely, that there is an interplay of temperature and time in the appearance of two differing kinds of behavior:

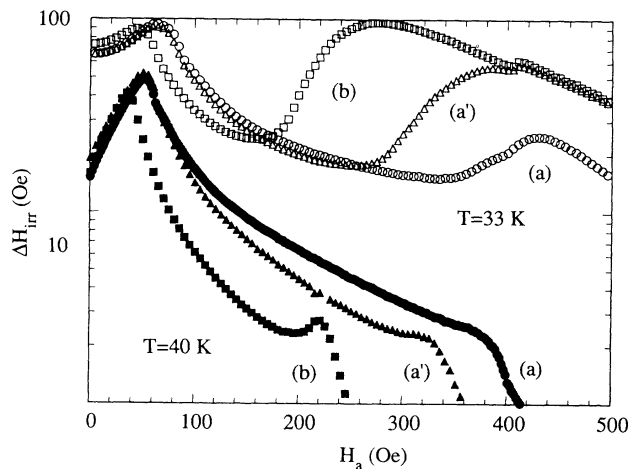


FIG. 2. Hysteresis-loop width  $\Delta H_{irr}$  vs applied field  $H_a$  along the  $c$  axis of BSCCO-2:2:1:2 crystals (a) as grown, (a') electron irradiated at  $4.4 \times 10^{18}/\text{cm}^2$  (same crystal), and (b) electron irradiated at  $2.2 \times 10^{19}/\text{cm}^2$  (different crystal). Both data sets show a sharp flux penetration at 70–90 Oe. The 33-K data show a second peak while the 40-K data show a step in field.

At short times in both data sets, there is a peak as a function of field, evident by taking a vertical cut through the data in Fig. 3 and appearing much as in Fig. 2 (33 K). The data at long times and lower current densities are more weakly time dependent, and at 35 K and long times, the peak of  $\Delta H_{irr}$  versus applied field has completely disappeared in favor of a step. At 31 K and long times, there is still a peak, but the slopes of the data suggest that at yet longer times, with higher-field curves falling more rapidly, the peak will also disappear. Thus the  $\Delta H$  peak is a dynamic phenomenon, and in this crossover region it can be made to disappear completely simply by waiting long enough.

The relaxation in most of this crossover region is best described by power laws of the form  $\Delta H = At^{-n}$ . This type of relaxation reflects a power-law voltage-current characteristic  $V \propto J^a$  which can be reconstructed by plotting  $\partial \Delta H / \partial t \propto V$  vs  $\Delta H \propto J$ . Values of the exponent  $a$  deduced in this way from the long-time data at 35 K, for example, drop almost linearly from 14 at 150 Oe to 5.5 at 350 Oe, with a rather sharp break to a lower, approxi-

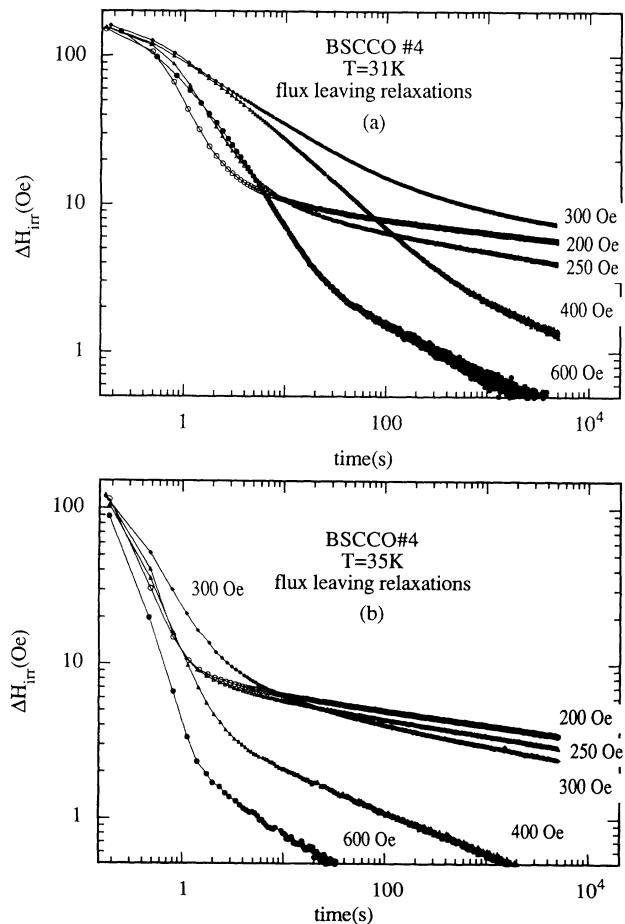


FIG. 3. Time decay of the irreversible hysteresis-loop width  $\Delta H_{irr}$  at (a) 31 K and (b) 35 K. These data imply a field peak at short times and a field step at long times.

mately linear falloff at higher fields, reaching 3.5 at 650 Oe.

These initially complex-looking data have a simple qualitative interpretation. Figure 3 suggests that at short times there is a source of large critical current density with strong pinning. However, it has a weak barrier to flux creep, causing this contribution to the current density to decay quickly. As the current density drops to low levels, a new mechanism of current density becomes evident, with a much higher barrier to flux creep causing slower relaxation.

We attribute the first, high-current-density mechanism to bulk pinning. This is supported by our observed enhancement of the hysteretic magnetization by irradiation (see Fig. 2) in this regime. The most direct evidence for this interpretation comes from several studies [18,19] which show proportionality of the hysteretic magnetization to crystal size as predicted by the Bean model for bulk pinning. These studies include one by Kishio *et al.* [20] on BSCCO crystals from the same source as ours.

The more novel point concerns the second, low-current-density regime, which we attribute to a surface barrier of the Bean-Livingston type [9-12] on the following basis.

(1) A fingerprint of the surface barrier is the characteristic asymmetric shape of the hysteresis loop in Fig. 1, which shows a sharp dropoff above flux penetration on the field-ascending branch, and a relatively flat portion close to  $H_s = 0$  on the higher-field portion of the field-descending branch. The  $H_s = 0$  behavior was first identified by Campbell and Evetts [21] as specific to a Bean-Livingston surface barrier: It comes from the fact that barriers to flux exit, normally arising from the surface current, disappear when the surface magnetization current of a uniform distribution of vortices is exactly canceled by the Meissner current induced by the applied field. Another example with  $H_s$  even closer to zero at somewhat higher temperature was shown earlier for one of our BSCCO-2:2:1:2 crystals [13].

We attribute the slight deviation from  $H_s = 0$  to some residual bulk pinning which increases at low fields, leaving a positive remanence. A possible explanation for the low-field increase is that even though bulk pinning almost vanishes for field parallel to the  $c$  axis in these highly two-dimensional materials at high temperatures, it could persist in the same temperature range for vortices lying along the  $a$ - $b$  planes; this in-plane component arises at low fields from curving of vortices into the plane because of demagnetizing effects [16,22].

The shape of the field-ascending branch has been predicted by Clem [23] to have the form

$$M = (H^2 - H_p^2)^{1/2} - H, \quad (1)$$

where  $H_p$  is the penetration field (e.g., 23 Oe in Fig. 1). A fit by this equation, taking  $H_s$  proportional to magnetization  $M$ , is shown by the solid line in Fig. 1. It should be recognized, however, that the applied field is oriented

perpendicular to the flat plane of the crystal, and the resulting demagnetizing effects have yet to be included in the theory.

(2) A second piece of evidence for the surface barrier interpretation is the decrease in current density due to irradiation shown in Fig. 2. Since irradiation introduces defects into the surface, it is likely to depress the surface barrier, as pointed out earlier in the Y-Ba-Cu-O context by Konczykowski *et al.* [12]. Related evidence of this contrast of low-temperature enhancement versus high-temperature suppression by irradiation can be found in the recent work by Shiraishi, Kazumata, and Kato [24] (see their Table I) although another study by Terai *et al.* [25] shows irradiation enhancement to much higher temperatures. We assume the crossover temperature depends on the specific bulk and surface perfection of each crystal and so may vary from study to study.

(3) A third piece of evidence of a surface barrier in the high-temperature regime is the lack of proportionality of  $\Delta H_{irr}$  with crystal size, reported in the same family of BSCCO-2:2:1:2 crystals by Kishio *et al.* [20]. Typically this effect has been attributed to granular behavior [20], but the surface-barrier interpretation is the only one consistent with the loop shape discussed above. It is also hard to accept that granularity could be given such a well-defined crossover observed as a function of time in Fig. 3 or as a function of temperature (e.g., see Refs. [6,7]).

An important result of our work is that each of the two regimes (bulk and surface-barrier) independently has its own complex dynamics and crossovers. For example, the fact that there is a peak in  $\Delta H_{irr}$  as a function of field at  $T = 33$  K, as shown in Fig. 2, indicates two competing mechanisms, and yet this peak falls entirely in the short-time, low-temperature regime of relaxation, just as in the short-time data of Fig. 3(a). Here we differ fundamentally from Kopylov *et al.* [11] who attributed the peak to the interplay of bulk and surface-barrier effects. We speculate that the  $\Delta H_{irr}$  peak is rather due to the interplay of some collective flux-creep barrier which increases with field, and the onset of a bulk irreversibility line caused by melting or reduction of the shear modulus of the vortex lattice.

More surprising is the fact that the surface-barrier regime has its own dynamic crossover. This is demonstrated by the change in the field dependence of the power-law exponent  $\alpha$  described above; this occurs at approximately the same field of 350 to 400 Oe where the hysteresis data show a step in Fig. 2. One can speculate that as field increases, the thermal activation over the surface barrier changes character, perhaps when the vortex spacing equals some defect spacing in the surface layer. Also of interest is the fact that surface-barrier relaxation is not simply logarithmic but shows a more complex power-law behavior. A first step in treating the dynamics of such barriers has been taken recently by Koshelev [26].

Clearly, studies which have ignored the crossover be-

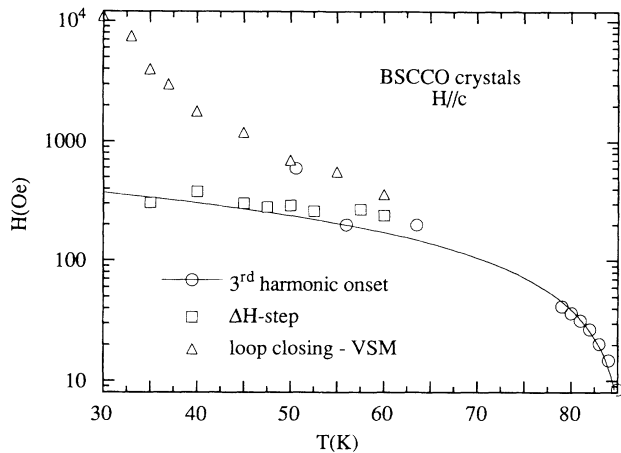


FIG. 4. Field-temperature plot of transition lines in BSCCO-2:2:1:2 crystals. Triangles represent the irreversibility line (loop closing) determined from vibrating-sample magnetization (VSM) measurements, squares represent the field step as in Fig. 1, and circles represent the onset of third-harmonic determination of the irreversibility line.

tween those two regimes need to be reexamined, especially where bulk-pinning concepts have mistakenly been applied to the high-temperature regime. An important insight can immediately be gained into the irreversibility line, in which de Rango *et al.* [27] early on pointed out the difference between high- and low-temperature character. We illustrate such data taken on our sample in Fig. 4. We have used a third-harmonic technique which will be described elsewhere but which essentially detects the deviation from linear response to ac-field amplitude in a magnetic susceptibility measurement [28]. The smooth extrapolation of the high-temperature irreversibility line to the long-time field step which we have reported in this paper suggests that the high-temperature portion should be interpreted in terms of the surface barrier, while de Rango *et al.*'s rapidly increasing portion at low temperature is a bulk effect.

In conclusion, this paper presents evidence for a crossover from a bulk pinning to a surface-barrier regime of current density. The surface-barrier regime is revealed to have its own characteristic dynamic crossover as a function of field. Surface-barrier effects are of interest for applications as well because they open up a new mechanism for current density in the high-temperature region where bulk pinning becomes weak in the BSCCO materials.

In the work of Weir *et al.* [29] on YBCO crystals, different flux-creep regimes were interpreted in terms of a surface-to-bulk crossover, but our result in BSCCO crystals differs substantially in the identification of these regimes.

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