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Surface flux pinning in $YBa_2Cu_3O_{7-\delta}$

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We report on the temperature and field dependence of the ac susceptibility and magnetic hysteresis of a number of $YBa_2Cu_3O_{7-\delta}$ single crystals with very low twin boundary densities and with defects added in a controlled manner to the crystal surfaces. It is shown that surface damage is by far the most important source of flux pinning, and that substantial shifts in the irreversibility line can be controlled by the number and type of defects added to the surface.

The many interesting features of vortex dynamics in $YBa_2Cu_3O_{7-\delta}$ single crystals have prompted a variety of theories of vortex states and possible transitions between these states.[1] The most dramatic transitions occur around a band of the magnetic field-temperature (H-T) plane usually called the irreversibility line (IL).[2] The irreversibility line $H_{irr}(T)$ divides the H-T plane into two regions: one at high fields and temperatures where neither flux pinning[2,3] nor a surface or edge barrier is active,[4–8] and the magnetization is reversible, and the other at low fields and temperatures where at least one of these mechanisms is active, yielding a nonzero critical current and hysteretic magnetic response. Initial understanding of the IL, and the associated changes in vortex dynamics, was developed in the context of the then available $YBa_2Cu_3O_{7-\delta}$ crystals which were dense with bulk pinning defects, especially twin boundaries. It was shown that in densely twinned $YBa_2Cu_3O_{7-\delta}$ single crystals, the presence of the twins completely dominates the magnetic ac response.[9] Refinement of crystal growth methods, and the development of stress annealing technology[10] to remove twin boundaries has resulted in nearly defect-free, untwinned crystals, which allow for more detailed investigation of both weaker types of pinning and of surface barrier effects. Resistivity measurements on such crystals have shown that the critical current suddenly disappears at a sharp boundary, associated with first-order vortex-lattice melting.[11,12] Since these crystals do not show any observable defects, the origin of the underlying pinning remains elusive, although it was shown by Konczykowski *et al.* that at temperatures close to T_c, the only source of hysteresis is a surface barrier opposing flux entry.[6] The barrier could be suppressed by low-temperature irradiation with 2.5 MeV electrons. A third mechanism, the pinning of magnetic flux by surface damage, has not yet received any attention.

This study probes surface flux pinning in $YBa_2Cu_3O_{7-\delta}$ crystals in the limit of extremely weak bulk pinning and correspondingly low disorder in the crystal structure. The positions of the H-T irreversibility lines were determined using ac inductance measurements, and were correlated with the shape of the magnetic hysteresis loops. The most uniform, defect-free crystals examined exhibit an IL that is governed by a surface, or edge barrier rather than pinning defects. Several different treatments are used to show the effects of surface damage on the IL in the H-T plane. Although the change in surface geometry could possibly affect the barrier against flux entry, it is shown that it is mainly the flux pinning that is considerably enhanced. In particular, in the limit of very weak bulk pinning and strong surface pinning, the IL shifts to high fields, and can occupy a position in the H-T plane that is normally associated with strong bulk pinning.

In the experiments reported here, $YBa_2Cu_3O_{7-\delta}$ crystals in as received condition were first measured to determine their initial IL's. In the first surface treatment, the crystals were placed in an iodine/ethanol solution and allowed to etch for 30 minutes. The crystals were then rinsed in ethanol and dried. Using scanning electron microscopy, this procedure was found to clean the crystal surface, making it smoother but without making any visible etch pits. A second treatment was to roughen the crystal surface by abrading it. Two methods of abrasion were used: one procedure was to scratch a controlled number of individual lines across the surface of

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the crystal with a sapphire needle. The second abrasion procedure was to lightly abrade the entire surface of the crystal with No. 900 carborundum grit held in silicon grease on a cotton tip. Other types of etchants are known to produce etch pits, hillocks, and screw dislocations in $YBa_2Cu_3O_{7-\delta}$. To test whether these defects might also prove to be flux pinning points, one crystal which had been previously etched as described above was additionally etched for 2 hours in glacial acetic acid. This etch condition has been shown to produce dislocation etch pits of 5–10 μm in diameter in $YBa_2Cu_3O_{7-\delta}$.[13]

The position of the IL was determined using ac inductance techniques described previously,[14] after each step in sample treatment. Briefly, a sample is mounted inside a mutual inductance coil set with the crystal c axis parallel to the ac and dc magnetic fields; the secondary coil voltage is proportional to the sample susceptibility, whose real (χ') and imaginary (χ'') components are measured with a phase-sensitive detector. χ'' is a measure of the ac magnetic field energy absorbed in the sample. A peak in χ'' occurs when the energy absorption is maximum, and is determined by the competition between the rise in the sample resistivity ρ due to vortex motion and the rapid fall of the shielding current density j upon increasing temperature. It has been shown[9] that the peak may indicate the appearance of flux pinning, which, while reducing ρ by opposing vortex motion, also prevents access of flux to an increasingly large part of the sample and thus increases the shielding current. The IL was determined as the locus of χ'' peaks. Its position in the H-T plane depends on the temperature and field dependence of the bulk shielding current density j, and is determined by the criterion $j[T,B(H),f]=h_{ac}/t$ (h_{ac} is the ac field magnitude and t is a sample dimension, in $YBa_2Cu_3O_{7-\delta}$ crystals typically the thickness). If there is more than one type of flux pinning present, the measurement of the ac susceptibility can result in more than one χ'' peak, each peak corresponding to the coincidence of an ac field penetration length with a typical length of the sample. For example, in the case of dilutely twinned $YBa_2Cu_3O_{7-\delta}$ crystals, the two peaks[9] correspond to the shielding of certain areas of the crystal due to the presence of the twin network, and to the shielding of the complete volume. The measurement of the IL thus allows the introduction or removal of different irreversibility mechanisms to be monitored systematically.

In the case of a surface or edge barrier a χ'' peak occurs when h_{ac} approximately equals the surface shielding current times the sample thickness. In order to distinguish between pinning, which acts against both flux entry and flux exit, and the surface barrier, which only opposes flux entry, we have also measured, using a superconducting quantum interference device magnetometer, magnetic hysteresis curves on one of the samples after each treatment.

Results of the experimental data are shown in Fig. 1 for a $650\times600\times30$ μm^3 typical $YBa_2Cu_3O_{7-\delta}$ crystal (JF31.26). The crystal had only a few twinning planes in one corner; as a result, the crystal had one main "lower" IL, with a very weak "upper" IL (not shown) at field/temperature values characteristic of pinning by twin planes.[9] As shown in Fig. 1, the initial iodine/ethanol etching treatment lowered the IL compared to the original data, which means that pinning centers were removed from the surface. The subsequent scratching of two lines in one face of the crystal did not produce a measurable change in the IL. However, the scratching of four additional lines in the same face of the crystal produced an IL higher than the original of the untreated crystal. Because of the smallness of the crystal, about 0.50 mm on a side, it was not possible to accurately orient the direction of the scratches. Finally, the whole face of the crystal that had been scratched was lightly abraded as described above. As shown in Fig. 1, on rerunning the sample, the IL was now much higher than before, approaching the slope produced by twin planes (crystal D357 in Fig. 1). Other crystals treated as above produced qualitatively similar results, i.e., weakly etching the surface typically lowered the IL, and abrading the surface raised the IL. Even though the degree in each case was sample dependent, the results clearly show the effect of the surface damage to be the addition of strong pinning centers. The same effects are shown in Fig. 2 for an

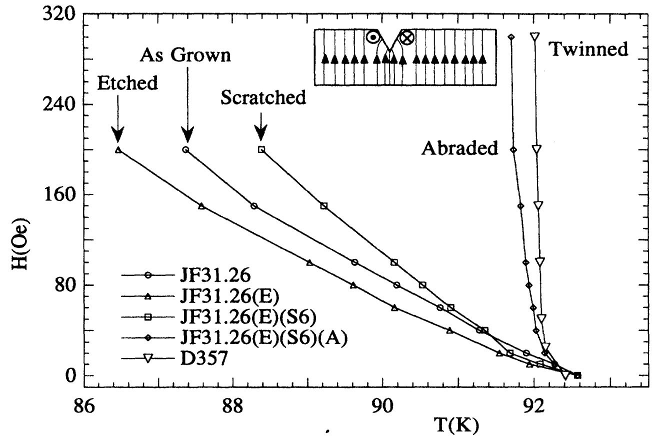

FIG. 1. The irreversibility line of $YBa_2Cu_3O_{7-\delta}$ sample JF31.26 in as grown condition (○), after etching (△), after scratching (□), after abrading (◇), and the IL of a twinned crystal (D357) (▽). The inset shows a schematic of the vortex and current configuration around a scratch.

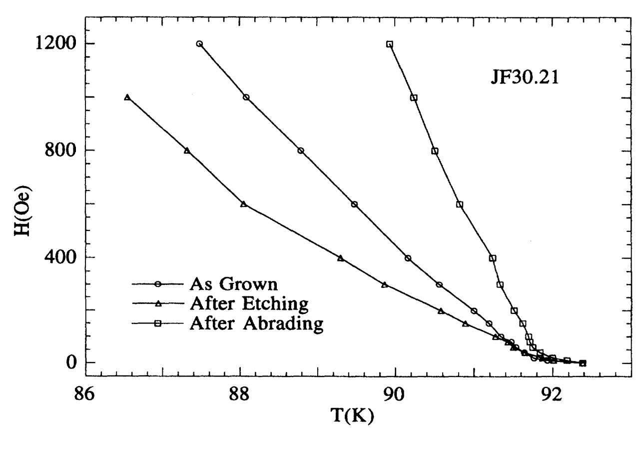

FIG. 2. The irreversibility line of $YBa_2Cu_3O_{7-\delta}$ sample JF30.21 in as grown condition (○), after etching (△), and after abrading (□).

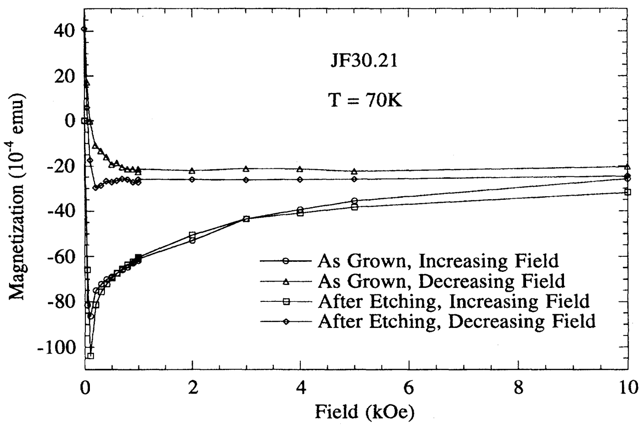

FIG. 3. Hysteresis loops for $YBa_2Cu_3O_{7-\delta}$ sample JF30.21 before (○) and after (□) an iodine/ethanol etch. The asymmetric hysteresis demonstrates the role of a barrier current.

$850\times500\times90$ μm^3 crystal JF30.21, which in its as-grown state was very similar to JF31.26. As before, the iodine/ethanol etch produced a lowering of the IL from the original, and abrasion raised the IL considerably above the sample in its original state.

The simultaneous change in the magnetic hysteresis is shown in Fig. 3 for crystal JF30.21, at a temperature of 70 K. A comparison of the results obtained on the original and the etched sample shows that the etch causes a decrease of magnetic hysteresis. Moreover, the hysteresis loop obtained after the etch shows the characteristic asymmetric shape associated with the presence of a surface barrier against flux entry only. This result means that any pinning centers initially present in the sample (apart from the few twin planes) were situated at the surface, and that these are completely removed by the iodine/ethanol etch. To underline this point, Fig. 4 shows the result of lightly abrading the whole sample surface: there is a significant increase in magnetic hysteresis, both in increasing and in decreasing field. The inset shows

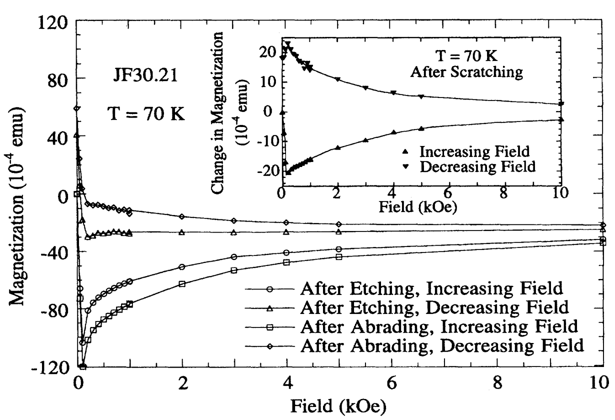

FIG. 4. Hysteresis loops of $YBa_2Cu_3O_{7-\delta}$ sample JF30.21 following an iodine/ethanol etch (○) and following light abrasion of the sample top surface (□). Inset: Difference in magnetic moment caused by scratching.

the difference between the magnetization of the sample before and after abrading. This is symmetric about zero, indicating that the extra hysteresis is the effect of increased pinning. It also means that the barrier was unchanged after abrading, for any change in the surface screening current would also have produced an asymmetry in the hysteresis difference.

The present result is in contrast to other work,[6] where it was shown that damaging of the surface by electron irradiation weakens the surface barrier and reduces magnetic irreversibility. The authors[6] concluded that the surface barrier was of the Bean-Livingston type.[4] This barrier arises as a result of the competition between the attractive force between a vortex inside the superconductor and its image outside, and the repulsion between the vortex and the surface screening currents. Surface irregularities on the scale of the magnetic penetration depth λ are thought to weaken both the attractive force of the image vortex and the repulsion by the surface currents, and therefore lower the Bean-Livingston barrier. The fact that the typical width of the scratches, $W=5$ μm, considerably exceeds the penetration depth for $YBa_2Cu_3O_{7-x}$, $\lambda(0)\approx120$ nm, means that the surface irregularities introduced here do not change the roughness on a small enough scale to affect the surface barrier. Another reason for the scratches not affecting the surface barrier may be that whereas the scratches were introduced on the top of the crystal, the main vortex penetration would be from the sides and edges, which did not undergo any change.

From the above discussion, we conclude that the main effect of the surface scratches is to introduce pinning sites. We estimate the pinning energy per unit length of a scratch as follows. A vortex line can gain energy $\epsilon_0 d$ (with $\epsilon_0=\phi_0^2/4\pi\mu_0\lambda^2$) by being inside a scratch of depth d. Comparing the width W of the scratch with the values of the vortex lattice parameter in our field range, 0.14 μm $<a_0<1$ μm, one sees that there are actually many vortices across the width of a single scratch. Extra vortices can take advantage of the energy gain in the scratch, but only by inducing a vortex lattice compression and tilt around the scratch. This will cause a local depletion of vortices near the edge just outside the scratch, and the appearance of an edge current (inset to Fig. 1).[15] The average optimal vortex displacement u_0 is obtained by comparing the energy of elastic deformation and the pinning energy, $c_{44}(u/W)^2dW \approx \epsilon_0 d(u/a_0^2)$, in the limit where W is much less than the typical sample dimensions. If $d\gg\lambda$ one should take $c_{44}=c_{44}(0)\approx B^2/\mu_0$, the local vortex lattice tilt modulus, then, $u_0\approx W(B_{c1}/B)$. The corresponding pinning energy per unit length is

$$U_p\approx\epsilon_0 d\frac{W}{4\pi\lambda^2}. \quad (1)$$

This energy associated with the depinning of the vortex lattice is very high. Moreover, there is no barrier preventing the vortices from reentering the scratch, so that thermal activation will not play an important role in depinning. Rather, the process of depinning will be very similar to the way in which the edge-shape barrier of Ref. 7 will be overcome. The critical current should be comparable to the edge current, and can be estimated from $U_p=Bj_sWdu_0$:

$$j_s = \frac{\epsilon_0}{\phi_0 W} \approx \frac{H_{c1}}{W \ln\kappa} . \tag{2}$$

An estimation of the position of the IL is complicated by the fact that the motion of flux around the scratch also has to be taken into account. A comparison of the different IL's shows that the area of the surface damage seems to be the controlling factor in the amount of pinning. The total length of the six scratched lines was about 2.6×10^3 μm, and the crystal area was about 3.5×10^5 μm^2. The ratio of $H_{irr}(T)$ for the whole area abraded sample and $H_{irr}(T)$ for the scratched sample is about 24. The scratched lines were measured to be about 5 μm wide, giving a ratio of the whole surface area to the area of the scratches of about 25, agreeing with the change in slope of the irreversibility line. Other observations have been made of this rough proportionality between the slope of the IL and the fraction of the surface area pinning flux. The surface damage in the final abraded state was much less extensive in Fig. 2 (crystal JF30.21) than in Fig. 1 (crystal JF31.26), leading to a lower, maximum slope for the IL line observed in Fig. 2. Other samples, in which etch pits have been produced,[13] show behavior similar to that in Fig. 2, with an upward, but less dramatic shift in the IL line. Work is in progress to try to identify the effective pinning area for defects other than scratches.

Lastly, since peaks in χ'' are directly related to changes in resistivity, it should be possible to correlate direct measurements of sample resistivity with the magnetization and susceptibility results. Lightly twinned $YBa_2Cu_3O_{7-\delta}$ crystals with an IL at a similar position in the (H,T) plane have shown a critical current "peak effect" and first order vortex melting.[16] Vortex lattice melting is characterized by a sharp jump in the resistivity, which signals the sudden disappearance of what was assumed to be bulk flux pinning. The present work shows that at high temperature, bulk pinning in these crystals is very weak. While this implies the degree of order in the vortex lattice is very high, it also means the role of surface imperfections should be considered in the interpretation of current experimental results.

In summary, we have shown that the main source of flux pinning in clean untwinned $YBa_2Cu_3O_{7-\delta}$ is imperfections and damage at the sample surface. The extent of the damaged surface area determines the slope of the irreversibility line; sufficiently important damage can move the IL to the position of the IL in densely twinned samples. The only other source of magnetic hysteresis at temperatures 86.0 K $<T<T_c$ is probably the edge barrier recently discussed elsewhere.[7,8]

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