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Evidence for surface barriers and their effect on irreversibility and lower-critical-field measurements in Y-Ba-Cu-O crystals

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We present measurements of the lowest field for flux penetration of untwinned $YBa_2Cu_3O_7$ crystals near their transition temperature T_c. This field decreases linearly with increasing temperature but exhibits an anomalous change in the slope in close vicinity of T_c. Low-dose electron irradiation reduces dramatically the first field for flux penetration. We discuss these results in terms of Bean-Livingston surface barriers which are proposed to be the origin for retardation in flux entry and for irreversible phenomena in the unirradiated crystals.

A reliable determination of the lower critical fields H_{c1} in high-temperature superconductors (HTSC) is still a challenge for experimentalists. The results of various experiments[1-6] agree only within a factor of 4–5, and also there are conflicting results[2,3,5,6] with regard to the temperature dependence of H_{c1}. In this paper we present an attempt to determine H_{c1} near T_c, for field parallel to the c axis, in *untwinned* $YBa_2Cu_3O_7$ crystals. In these crystals, unlike the situation in the twinned ones, first flux penetration can be easily detected via a sharp kink in the magnetization curves. In addition, we improve the sensitivity of this experiment to the level of a single-flux-line detection, by using a miniature Hall probe. The combination of the sharp kink and the improved sensitivity allows us to determine the first field for flux penetration with very high accuracy. The results of these measurements exhibit an anomalous change in the apparent slope dH_{c1}/dT in the vicinity of T_c. This anomalous feature is sample dependent and can be suppressed by electron irradiation. Moreover, the measured first field for flux penetration is much larger in the sample before irradiation, clearly demonstrating the existence of barriers to flux penetration which, presumably, are responsible for the wide spread in the experimental $H_{c1}(T)$ data.

We present data for three untwinned samples of dimensions $1090\times460\times4$ and $1000\times460\times4$ μm^3 (which are two pieces of what was originally one crystal) and $1700\times1400\times23$ μm^3. We refer to these samples as U1a, U1b, and U2, respectively. The details of crystal growth are given in Ref. 7. The method uses an off-stoichiometric composition rich in $BaCuO_2$ and CuO. Samples are grown in gold crucibles in air and post annealed at 420 °C in oxygen for ten days. For comparison, we discuss briefly our results for twinned samples; crystal growth is described in Ref. 8.

Irradiation at low temperature (20 K) by 2.5 MeV electrons from a Van de Graaff accelerator produced randomly-distributed isolated Frenkel pairs. The damage produced by fluence of 0.138 C/cm^2 is estimated[9] to be 3×10^{-4} d.p.a. (displacement per atom). Agglomeration of small defect clusters is expected when the sample is warmed to room temperature for transfer to the measuring cryostat. The remaining damage, after room-temperature annealing, is in the range of $(0.6-1.2)\times10^{-4}$ d.p.a.

The key technical point of our measurement method is the use of *a miniaturized* InSb Hall sensor[10] with 80×100 μm^2 active area, of highly linear slope (typically 50 mΩ/G) which is almost temperature independent below 120 K. The probe is used for measurements of magnetization as a function of temperature, field, or time with a resolution of 0.004 G, corresponding to approximately one flux line on the probe surface. An external field H_a is applied and the field H_m measured by the probe is detected. As a result of perturbation, by the sample, of the homogeneous applied field $H_m \neq H_a$; we define $\Delta H = H_a - H_m$. This perturbation ΔH is related directly to the magnetization of the sample; henceforth we refer to it as the magnetization field. In Fig. 1(a) we demonstrate the technique by presenting magnetization curves for sample U1a before irradiation. The initial linear slope is related to the Meissner state. The actual slope of ΔH vs H_a in this region depends on the position of the Hall probe.[11] It is important to note, however, that we find no effect whatsoever of the location of the probe on the first field for flux penetration, denoted by the kink in the magnetization curve.[10]

For a first characterization of the three samples we performed conventional zero-field-cooled (ZFC), field-cooled (FC), and thermoremanent magnetization measurements between 80 and 95 K. The ZFC-FC study yields a much wider reversible regime for the untwinned samples. The thermoremanent magnetization study shows that the twinned samples trap practically all the field H_a to which they were exposed during the cooling process, whereas the untwinned crystals exhibit extremely small remanence. These two observations imply that the untwinned samples

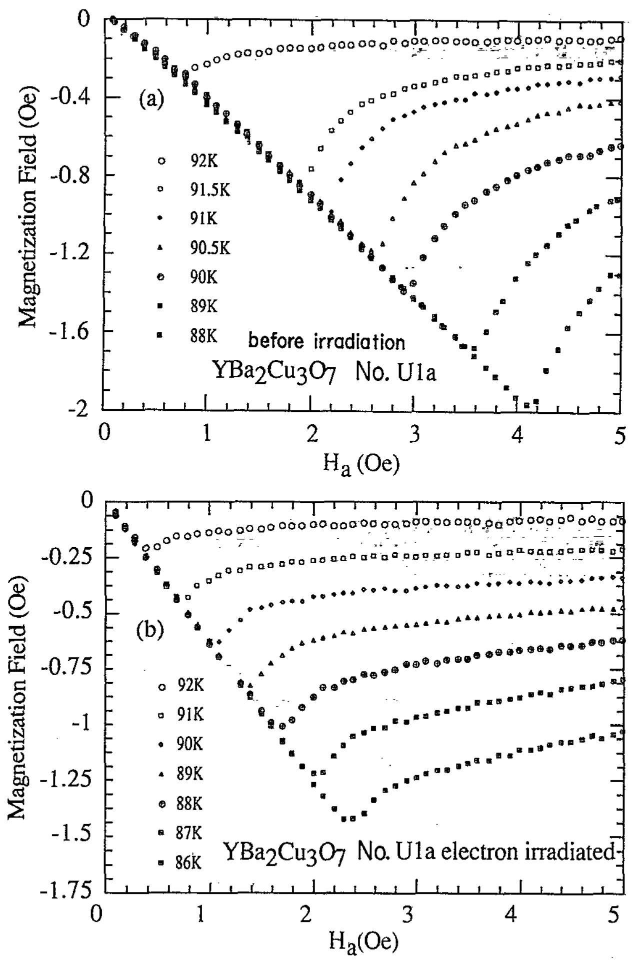

FIG. 1. Magnetization curves for untwinned Y-Ba-Cu-O, sample U1a, at the indicated temperatures (a) before and (b) after irradiation.

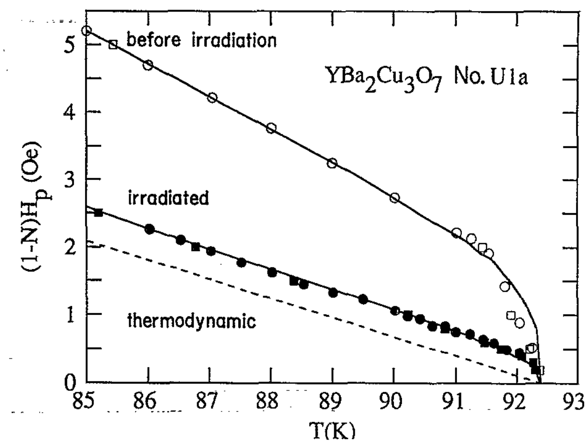

FIG. 2. Temperature dependence of H_p, the first field for flux penetration, for untwinned sample U1a before and after irradiation. Circles refer to points deduced from magnetization curves and from the onset of remanent magnetization. Squares present data of ZFC magnetization. Open and solid symbols refer to measurements on sample before and after irradiation, respectively. The solid lines are guides for the eye. The dashed line is our estimation of the thermodynamic H_{c1}. Applied fields are described in this figure. Actual values can be obtained by dividing the quoted values by $1-N$.

are less efficient in trapping flux. This conclusion is further supported by the magnetization curves [Fig. 1(a)] of the untwinned samples. While the twinned sample exhibits the familiar behavior, namely a slow deviation from linearity and a broadening of the peak, the untwinned samples exhibit a sharp cusp at a well-defined field H_p, the first field for flux penetration. It is tempting to identify H_p as H_{c1}, the lower-critical field. However, we demonstrate later that energy barriers for flux penetration cause $H_p > H_{c1}$.

The temperature dependence of H_p was deduced from the kink in the magnetization curves. The actual value of H_p has to be corrected by a factor of $1-N$, N being the demagnetization factor. The values $N=0.97$ and 0.86 for samples U1a, U1b, and U2, respectively, are deduced from the initial slope of independent $M(H)$ measurements on a calibrated device (a commercial SHE susceptometer), assuming a perfect shielding ($=-1/4\pi$). Using the ellipsoidal approximation we get slightly higher values, implying that much larger corrections might be necessary. To avoid possible errors in demagnetization corrections, we show here only raw data. The temperature dependence of H_p for sample U1a is described in Fig. 2 by the open circles. Two features are apparent in this figure. (i) Initially, H_p drops linearly with increasing temperature; the line extrapolates to zero at $T_0=95.7$ K. A similar T_0 is found for samples U1b and U2. As described below, after electron irradiation T_0 is closer to T_c. (ii) At a sample dependent temperature T_b the slope dH_p/dT crosses over to the larger value and $H_p(T)$ drops to zero at $T_c=92.4$ K. The temperature $T_b=91.2$ K for samples U1a and U1b and 90 K for the *thicker* sample U2. We stress the fact that the thicker sample shows the anomalous break at *lower* temperature; this rules out explanations of this feature which are based on the geometry, for example, on the ratio of the thickness to the London penetration depth $\lambda(T)$.

For a cross check of the behavior described above, we remeasured $H_p(T)$ by two other independent procedures. The first procedure[2] is based on measurements of the ZFC magnetization curves as a function of temperature. In this technique, flux penetration is identified via a sharp drop in the diamagnetic shielding. Results for H_p from measurements of ZFC magnetization are described in Fig. 2 by the open squares. The second technique[4,5] is based on measurements of isothermal remanent magnetization as a function of the field: The sample is cooled in zero field, a field H_a is turned on and then off. The field H_p is identified via the onset of remanent magnetization. We find a sharp onset of remanence at exactly the same field as the kink in the magnetization curve. Thus, the three procedures yield similar values for $H_p(T)$.

Electron irradiation has a dramatic effect on the features described above. The electron-induced damage $[(0.6-1.2)\times 10^{-4}$ d.p.a.] of this experiment has no effect on T_c but the measured first field for flux penetration has been dramatically reduced. This is clearly demonstrated by comparing Figs. 1(a) and 1(b) which show magnetization curves before and after irradiation, for the same sample. Note that the kink in the magnetization curve is quite sharp and first field for flux penetration can still be well defined. The measured $H_p(T)$ values of the irradiated sample are described by the solid circles in Fig. 2. Identical results were obtained from the onset of isothermal remanent magnetization. Results of ZFC magnetization measurements are described by the solid squares in this figure.

Figure 2 summarizes the main effects caused by irradiation, namely, reduction in the apparent first field for flux penetration and suppression of the anomalous change of the slope of $H_p(T)$. The fact that the first field for flux penetration is reduced significantly in the irradiated sample suggests the presence of energy barriers for flux penetration in the unirradiated crystal. These barriers, which cause a retardation in the first field for flux penetration to fields $H_p(T) > H_{c1}(T)$, are destroyed by irradiation. Thus, the data for the irradiated sample are closer to the true thermodynamic behavior, which is shown schematically by the dashed line.

A clue to the origin of the barriers for flux penetration can be obtained from the analysis of the magnetization loops presented in Figs. 3(a) and 3(b). Two features are apparent in these figures: The *almost-zero magnetization* of the descending branch of $M(H)$ prior to irradiation and the significant *reduction*[12] *in the "width" of the hysteresis loops* of the irradiated sample. These features persist at *all* temperatures down to 70 K. Strong bulk pinning is expected to produce $M(H)$ loops which are symmetric around zero magnetization. Asymmetry in the magnetization loops is usually interpreted as a superposition of reversible and irreversible contributions. This explanation is unlikely to be valid for Fig. 3(a), due to the fact that M of the descending branch is almost zero over a wide range of temperatures. On the other hand, as pointed out by Campbell and Evetts,[13] almost-zero magnetization of the descending branch is one of the main fingerprints of the Bean-Livingston[14] (BL) surface barriers. According to BL, the barriers arise from the competition between two forces: (1) Attraction between the flux line and its mirror image, pushing the vortex line outside the sample. (2) Interaction between the line and surface shielding currents pushing it inside the sample. Shielding currents, being proportional to the magnetization field ΔH, go to zero at the early stages of the descending branch of the loop. Once $\Delta H \approx 0$ the BL barriers disappear; when the field is further reduced flux is able to leave the sample freely until the overall magnetization is stabilized again at this value.[13] This situation is completely changed after irradiation; the narrow width of the loop after irradiation reflects the depression of surface barriers. This is, of course, consistent with the decrease in H_p.

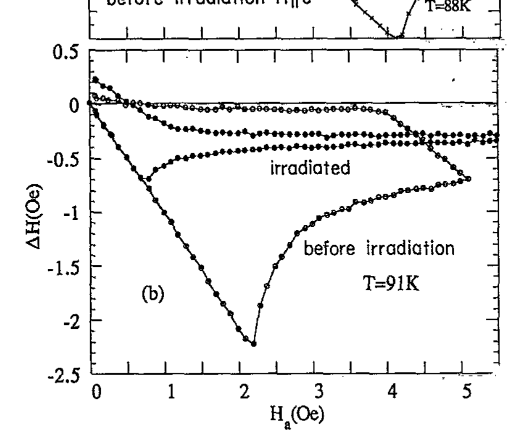

FIG. 3. (a) Typical magnetization loops for sample U1a before irradiation. Note the almost-zero magnetization of the descending branch. (b) Magnetization loops recorded at 91 K for sample U1a before and after electron irradiation. The initial Meissner slope was normalized to -1, by dividing each M value by apparent value of initial slope, in order to account for small difference of Hall probe position in two runs.

Suppression of BL barriers by surface modification has been observed in the past. The most relevant example is ion implantation in Mo-Nb which dramatically reduces the barriers.[15] The low-dose irradiation of the present experiment has no apparent effect on the surface of the sample. However, 2.5 MeV electrons are likely to produce surface damage because of migration of defects from the bulk to the surface during room-temperature annealing.[9] We therefore assert that damage induced by irradiation reduces the barriers, having an effect similar to that reported in the literature.[15]

Anomalous temperature variation of the first penetration field H_p near T_c can be tentatively accounted for in the following way. A necessary condition for the attractive force between the vortex line and its image to be effective is that the surface should be smooth over a distance comparable to λ. Near T_c, λ is larger than existing surface irregularities, thus BL barriers are fully effective. At lower temperatures λ is comparable to surface irregularities and BL barriers are less effective. Similar considerations of interplay between λ and surface roughness were presented in Ref. 16.

Finally, we comment on measurements on twinned samples. Several factors cause the spread of results of these measurements. We have already mentioned, in the introduction, that in twinned crystals onset of irreversibility is blurred because of flux pinning, hence, for example, the rounding of the magnetization curves. The most important factor affecting H_p is apparently the effect of the barriers on flux penetration. Such barriers have already been proposed[5] to explain the low-temperature anomaly in H_p and their existence is clearly demonstrated in this work. Also, though the influence of twins is still highly controversial[17] it is clear that the presence of twin boundaries complicates the effect of the barriers: In some circumstances, depending on the density and orientation of the twins,[18] the barriers may not be effective in preventing a vortex to pass through the surface inside the bulk along twins. In our own measurements on two twinned samples we find no anomaly in the temperature dependence of $H_p(T)$ for one sample and a kink, similar to that described in Fig. 2, for the second one. These results will be described elsewhere.

In conclusion, we presented evidence that Bean-Livingston surface barriers, not bulk pinning, are the origin of the apparent irreversibility in high quality $YBa_2Cu_3O_7$ single crystal near T_c. These barriers are probably responsible for the anomalous temperature dependence of H_p. It should be stressed that BL barriers would be particularly effective in HTSC (Ref. 19) due to their high value of the Ginzburg-Landau parameter κ. It is not clear to us to what degree these barriers affected previous results but it is obvious that the BL barriers are responsible for the wide spread of conflicting experimental data. Thus the present results suggest the need for reevaluation of earlier measurements of $YBa_2Cu_3O_7$ crystals.

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