PHYSICAL REVIEW B VOLUME 47, NUMBER 9 1 MARCH 1993-I

Evidence of surface barriers in single-crystal $Tl_2Ba_2CuO_6$ superconductors

F. Zuo and D. Vacaru
Physics Department, University of Miami, Coral Gables, Florida 33124

H. M. Duan and A. M. Hermann
Department of Physics, University of Colorado, Boulder, Colorado 80309

(Received 5 November 1992; revised manuscript received 8 December 1992)

We report direct evidence of surface barriers in single crystals of $Tl_2Ba_2CuO_6$ superconductors. Magnetization is measured as a function of temperature with magnetic field applied parallel to the c axis. For small field, a characteristic linear increase in M is observed at an onset temperature T_1. For higher magnetic field, a new feature at low T is observed in the $M(T)$ data. The flux penetration temperature is retarded to a higher temperature. The penetration field H_p at temperature below 50 K determined by the first linear rise in $M(T)$ can be fitted with the relation $H_p(T)=H_c\exp(-T/T_0)$, consistent with a model where two-dimensional pancakes penetrate the Bean-Livingston barrier by thermal activation. The low critical field $H_{c1}^{\perp}$ determined from the bulk penetration of vortices is also presented.

The evidence for Bean-Livingston (BL) surface barriers has been reported from studies of magnetization (hysteresis loops) in the high-temperature superconducting materials.[1-3] For untwinned Y-Ba-Cu-O crystals, the hysteresis loop is not symmetric and the magnetization M is almost zero in the descending branch; the magnetic penetration field is reduced upon electron irradiation rather than increased.[1] Magnetization studies on the $Bi_2Sr_2CaCu_2O_8$ and $Tl_2Ba_2CaCu_2O_8$ show that the penetration field $H_p(T)$ has a positive curvature at low temperature. The temperature dependence comes from the thermally activated hopping of vortices over the BL surface barrier.[2] We report here a direct evidence for the BL surface barrier from measurements of zero-field cooled (zfc) magnetization measurement in single crystals of $Tl_2Ba_2CuO_6$ superconductors. At high temperatures, the penetration field corresponds to the lower critical field H_{c1}. At low temperatures, the penetration of vortices is thermally activated, $H_p(T)=H_0\exp(-T/T_0)$. We interpret this as evidence that the vortices in this material are manifestly two-dimensional.

Single crystals of $Tl_2Ba_2CuO_6$ were prepared by a solid-state self-flux method.[4] The samples used in the experiments were platelets with average dimensions 0.8 mm×0.3 mm×0.04 mm. The magnetization measurements were performed with a Quantum Design superconducting-quantum-interference-device susceptometer. The crystal we discuss here had an onset transition temperature $T_c=88.8$ K and a transition width $\Delta T\approx 3$ K at $H=1$ G. Several samples were studied yielding very similar results. The remanent field of the superconducting magnet is normally less than 0.05 G after quenching the magnet. The samples were zero-field cooled (zfc) to a set temperature, and a magnetic field of 0.1 G to 1 T was applied parallel to the c axis. The magnetization was then measured with increasing temperature. For this configuration, the specimen can be approximated to an oblate disk with the aspect ratio of $0.5/0.04=12.5$, yielding demagnetization factor $1/(1-N)=8.8$.

Shown in Fig. 1(a) is a typical plot of low-field zfc magnetization versus T. The sample was zero-field cooled (<0.05 G) to 5 K and the measurement was performed in a constant field as the temperature was increased. The magnetization has been normalized to its low-temperature value M(5 K). A linear rise in M/M(5 K) is clearly seen in the data for $T\geq 60$ K, followed at higher temperatures by a more rapid increase toward $M=0$ at T_c. We define T_1 to be the temperature where the linear-T data extrapolates to the full Meissner effect or the M/M(5 K)$=1$ line. T_2 defines the temperature above which M deviates from the linear-T behavior.

The observation of T_1 has been reported in the zfc magnetization of Y-Ba-Cu-O and $Bi_2Sr_2CaCu_2O_8$ crystals.[5] The onset of the linear rise is associated with the bulk penetration of vortices into the sample, and the linear temperature dependence can be easily derived using Bean's critical state model. The extrapolation to the full Meissner effect gives the corresponding critical temperature in $H_{c1}(T)$ for a given applied field.[5,6] The $H_{c1}(T)$ measured at high temperature using this technique on Y-Ba-Cu-O crystal agrees well with BCS theory. It is important to note that the low critical field measured this way is more accurate than by the method of magnetization as a function of field at a given temperature, because it is determined only by the bulk penetration. The fast decay of $M(T)$ for $T>T_2$ may be due to flux depinning at high temperatures.

Figure 1(b) is a plot of zfc magnetization at $H=60$ G. Unlike the data at $H=20$ G, a new feature has emerged in the data. The magnetization rises suddenly at $T=T_p$ and converges to the linear-T dependence. The extrapolation of the linear high-temperature data intercepts with the $M(T)/M$(5 K)$=1$ line at $T=T_1$, which is less than the retarded penetration temperature T_p. T_p is defined as the intercept of the low-temperature line with the $M(T)/M$(5 K)$=1$ base line as shown in the graph. The vortices appear to be inhibited from entering the sample at $T=T_1(H=H_{c1})$. A higher temperature T_p is evidently required to enable the flux penetration.

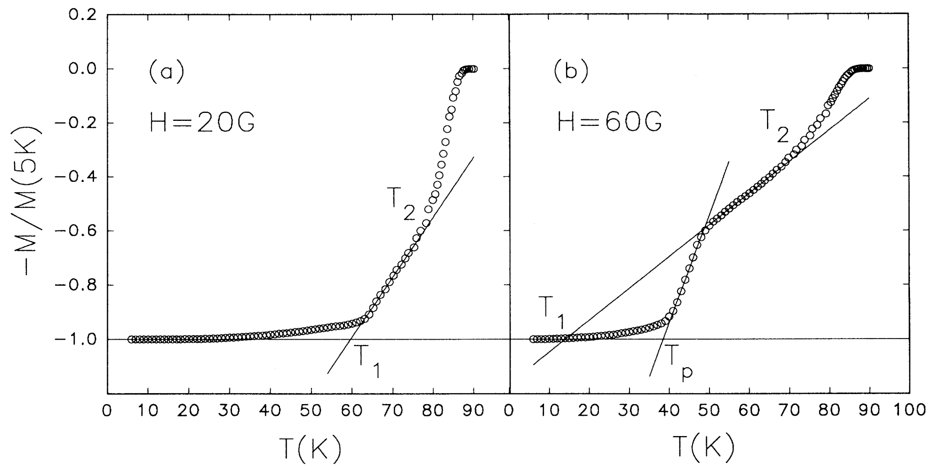

FIG. 1. The normalized zfc magnetization as a function of temperature: (a) $H = 20$ G; and (b) $H = 60$ G. The solid lines are guides for extrapolation.

Plotted in Fig. 2 are M versus T data for different applied fields $H = 15$, 20, 30, 45, 60, 75, and 100 G. At low fields ($H < 30$ G), the overall profile is the same as discussed above with characteristic temperatures T_1 and T_2. At higher applied magnetic field, one can see that the upturn at $T = T_p$ is more pronounced. We find $T_p = 48$, 44, 38, 35, and 30 K for $H = 30$, 45, 60, 75, and 100 G, respectively. The extrapolated T_1 approaches zero at $H = 75$ G.

The onset and the subsequent growth of retarded penetration at T_p in the zfc magnetization are direct evidence of surface barriers. Earlier measurements on BL surface barriers have been confined to the magnetization as a function of field at a fixed temperature. There, the penetration field is defined as the field where a deviation from a linear $M(H)$ is observed. Methods to extrapolate the intrinsic lower critical field have been discussed extensively.[1] The advantage of performing zfc magnetization and using the characteristic linear increase with T to obtain bulk penetration field is that this method eliminates possible artifacts due to small vortex leakage at the surface.[5]

Figure 3 shows the experimental results of H versus the extrapolated T_p and T_1. The upper data set corresponds to the $(1-N)H_p(T)$, and the lower data set to the $(1-N)H_{c1}^{\perp}(T)$. Clearly, $H_p(T)$ increases with decreasing temperature with a positive curvature in T. Only data with field up to 400 G are plotted. The sharp transition in higher applied field is smeared out, and it is difficult to draw the base line to find an extrapolation for T_p. $H_{c1}^{\perp}(T)$ shows a much weaker temperature dependence. At low temperature, $H_{c1}^{\perp}(T)$ is approximately linear with T, with a zero-temperature value of 75 G.

Shown in Fig. 4 is a plot of $\ln[(1-N)H_p]$ versus T. The solid line is a least-squares fit of the low-temperature data to $\ln[(1-N)H_p(T)] = a + T/T_0$, with $a = 6.9 \pm 0.2$ and $T_0 = 14 \pm 1$ K. A good fit is clearly seen between the data and the line.

The observation of T_p in zfc magnetization is a direct measurement of the penetration field in the presence of

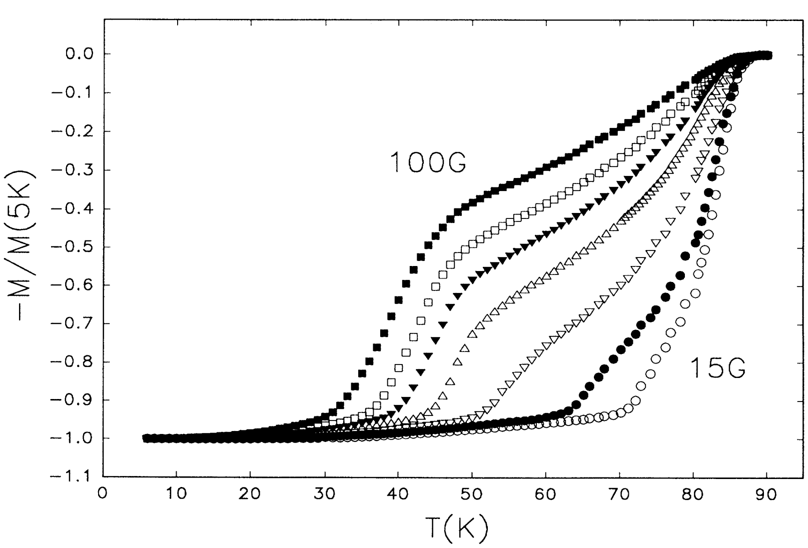

FIG. 2. An overlay of zfc magnetization as a function of field. The applied field $H = 15$, 20, 30, 45, 60, 75, and 100 G.

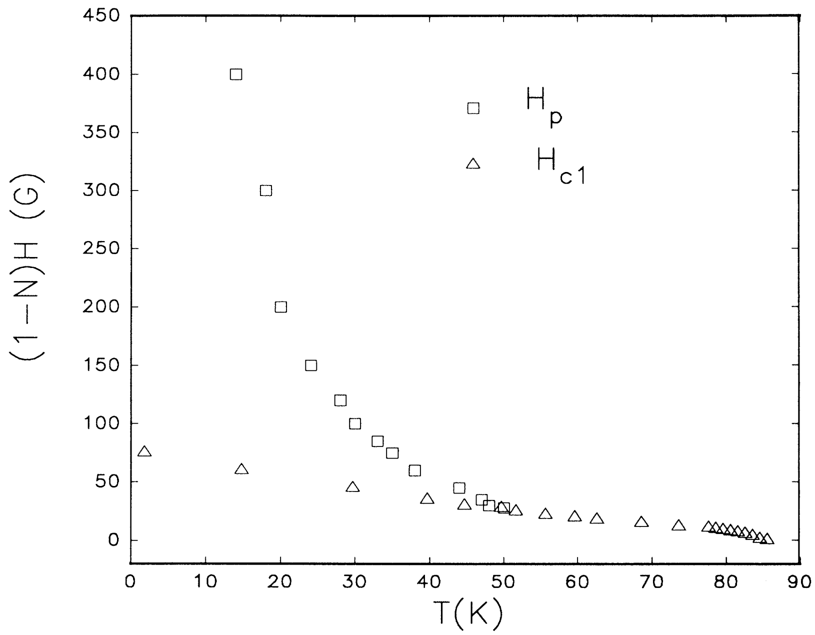

FIG. 3. Plot of the extrapolated $(1-N)H_{c1}^{\perp}$, and $(1-N)H_p$ as a function of temperature.

the BL surface barrier. The BL surface barrier arises from the competing effects of an attractive interaction of the Abrikosov vortex to its "mirror image" near the surface and the repulsive interaction on the vortex from the applied field.[7] The barrier can be reduced or neglected in cases where surface superconductivity is suppressed due to oxidation, surface roughness, and presence of defects, such that the penetration field measured is the lower critical field H_{c1}. In the presence of the BL surface barrier, the penetration field to overcome the surface barrier can be easily shown to be the thermodynamic critical field $H_c=(\phi_0/4\pi\lambda\xi)\ln(\lambda/\xi)$. For conventional superconductors, H_c can be well approximated by a parabolic law $H_c(T)=H_0[1-(T/T_c)^2]$.

The temperature dependence observed in $Tl_2Ba_2CuO_6$ is certainly inconsistent with the BCS theory, where one expects the critical field H_c to saturate at relatively low temperature, rather than to increase exponentially with decreasing temperature. It has been proposed that the thermal activation plays an important role in the penetration of two-dimensional vortices in the $Tl_2Ba_2CaCu_2O_8$ and $Bi_2Sr_2CaCu_2O_8$ compounds.[2] For conventional type-II superconductors, Bean and Livingston showed that for a semi-infinite sample with field parallel to the surface, the barrier energy per unit length was given by $U_b(H)=\phi_0^2/(4\pi\lambda)^2\ln(H_c/H)$.[7] For two-dimensional (2D) systems such as superlattices or thin films, one can assume the current distribution to be uniform (for

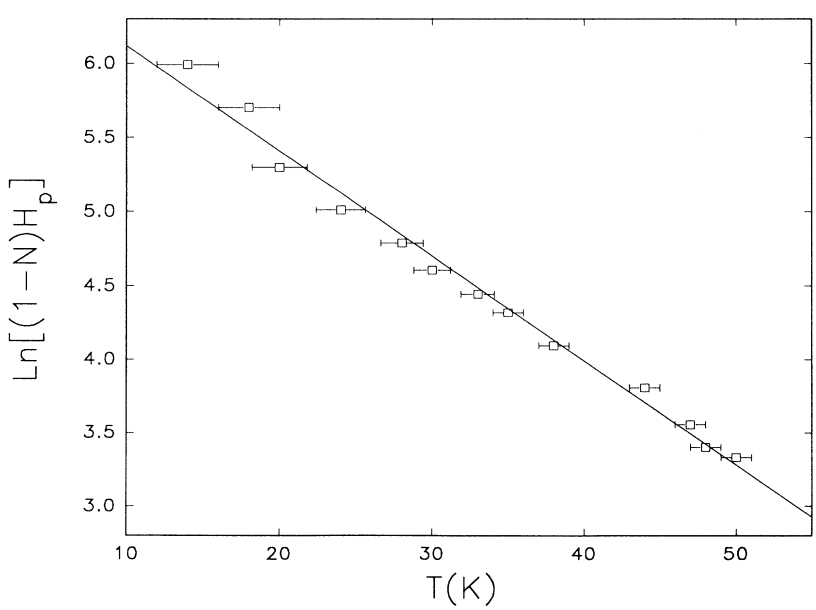

FIG. 4. $\ln[(1-N)H_p]$ as a function of temperature. The solid line is a fit to the data.

$d/\lambda \ll 1$) within the layer, such that the barrier height can be approximated by[2,8]

$$U_b(H)=\phi_0^2 d/(4\pi\lambda)^2 \ln(H_c/H) , \quad (1)$$

where ϕ_0 is the flux quantum, d is the thickness of the superconducting layer, λ is the penetration depth, and H_c is the thermodynamic critical field. At high temperature, the vortex penetration is limited by the bulk pinning. At low temperature, $k_B T < U_b$, vortices are thermally activated over the barrier. Qualitatively, one can obtain the temperature dependence of the penetration field by setting $k_B T = \alpha U_b(H)$, with $\alpha < 1$, then

$$H_p(T)=H_c \exp(-T/T_0) . \quad (2)$$

The exact form of T_0 is complicated due to the nature of the thermal activation. By considering the activation process and the appropriate time window for the measurement, it has been shown[2] that $T_0=\phi_0^2 d/(4\pi\lambda)^2 \ln(t^*/t_0)$, where t^* is the typical measurement time (~ 100 s), and $1/t_0$ is the hopping frequency, with $t_0 \approx 10^{-1}-10^{-11}$ s. If we take the layer thickness to be 4 Å, and $T_0=14$ K, we calculate $\lambda \approx 1400$ Å. To get another estimate for λ, we extrapolate the $H_{c1}(T)$ at low T to get $H_{c1}(T=1) \approx 600$ G. Using $H_{c1}=\phi_0/(4\pi\lambda^2)\ln\kappa$, where κ is the Ginsberg-Landau parameter (~ 100), we get $\lambda \approx 1100$ Å. The zero-temperature critical field H_c is about 9 kG after taking the demagnetization factor into account.

The experimental results fit well with a thermally activated penetration of 2D vortices in the $Tl_2Ba_2CuO_6$ compound. The thermal energy is important for 2D pancakes, but it is negligible for 3D flux lines, because the length involved in the barrier height is the dimension of the sample instead of the thickness of the layer. The observation of 2D vortices is consistent with the large anisotropy ($\gamma=\sqrt{M_c/M_{ab}} > 100$) measured from torque measurements,[9] and high field magnetization measurements on $Bi_2Sr_2CaCu_2O_8$ and $Tl_2Ba_2CuO_6$ compounds,[10,11] where experimental results were modeled with weak Josephson coupling between the superconducting layers.[12]

Another possible explanation for the positive curvature in $H_p(T)$ is the proximity effect which can yield a positive curvature in $H_{c1}^{\perp}(T)$ at low temperature depending on the anisotropy.[13] The basic idea is that as the temperature is lowered, the order parameter in the normal metal layer (N-layer) between the superconducting layers increases due to proximity effect. Some energy has to be spent in the creation of vortices in the N-layer, leading to an increase in $H_{c1}^{\perp}$ at low T. However, the positive curvature is obtained only when the N-layer is highly conducting, and the temperature dependence of $H_{c1}^{\perp}(T)$ is a strong function of the anisotropy γ. It is unlikely that the exponential dependence we observed comes from the proximity effect.

In summary, we have reported a detailed magnetization study on single-crystal $Tl_2Ba_2CuO_6$ samples. From zfc magnetization $M(T)$, we have clearly observed the onset and the subsequent growing effect of the BL surface barrier. The temperature dependence of $H_p(T)$ is consistent with a model where 2D pancakes hop over the BL barrier by thermal activation. This result supports the picture that the $Tl_2Ba_2CuO_6$ system at low temperature can be described as a weakly coupled layered superconductor.

We acknowledge many useful discussions with Dr. Stewart Barnes and Dr. Joshua Cohn. This work was supported by a general research grant of the University of Miami (F. Z.).

[1] L. Burlachkov, Y. Yeshurun, M. Konczykowski, and F. Holtzberg, Phys. Rev. B **45**, 8193 (1992); M. Konczykowski, L. I. Burlachkov, Y. Yeshurun, and F. Holtzberg, *ibid.* **43**, 13 707 (1991).

[2] V. N. Kopylov, A. E. Koshelev, I. F. Schegolev, and T. G. Togonidze, Physica C **170**, 291 (1990).

[3] V. M. Krasnov, Physica C **190**, 357 (1992).

[4] M. Parathaman, H. M. Duan, and A. M. Hermann, in *Thallium-Based High Temperature Superconductors,* edited by A. M. Hermann and J. V. Yakhmi (Marcel Dekker, New York, 1992).

[5] L. Krusin-Elbaum, A. P. Malozemoff, Y. Yeshurun, D. C. Cronemeyer, and F. Holtzberg, Phys. Rev. B **39**, 2936 (1989).

[6] T. Matsushita, E. S. Otabe, T. Matsuno, M. Murakami, and K. Kitazawa, Physica C **170**, 375 (1990).

[7] C. P. Bean and J. D. Livingston, Phys. Rev. Lett. **12**, 14 (1964).

[8] S. E. Barnes, J. Phys.: Condens. Matter **4**, 4135 (1992).

[9] D. E. Farrell, R. G. Beck, M. F. Booth, C. J. Allen, E. D. Bukowski, and D. M. Ginsberg, Phys. Rev. B **42**, 6758 (1990).

[10] P. H. Kes, C. J. van der Beek, M. P. Maley, M. E. McHenry, D. A. Huse, M. J. Menken, and A. A. Menovsky, Phys. Rev. Lett. **67**, 2383 (1991).

[11] D. Vacaru, F. Zuo, H. M. Duan, and A. M. Hermann, Physica C **203**, 393 (1992).

[12] L. N. Bulaevskii, M. Ledvij, and V. G. Kogan, Phys. Rev. Lett. **68**, 3773 (1992).

[13] A. A. Golubov and V. M. Krasnov, Physica C **196**, 177 (1992).