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Superconducting transport properties of epitaxial $YBa_2Cu_3O_{7-\delta}$ thin films: A consistent description based on thermally-activated flux motion

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The electrical-transport current properties of a series of epitaxial $YBa_2Cu_3O_{7-\delta}$ thin films have been investigated using a range of techniques, including the activated electrical resistivity for fields $B > B_{irr}$, the irreversibility field; the I-V curves for both $B > B_{irr}$ and $B < B_{irr}$; and the resistive transitions in magnetic fields. The results are analyzed in the framework of a model for thermally-activated flux motion. The model utilizes a pinning barrier that is dependent on the current density J, temperature T, and applied magnetic field B, given by $U_0 \propto \exp(-J/J_{c0})(1-t)^n/B$, with $n \sim 1.8$. The exponential J dependence agrees well with the behavior $U \propto J^{-\mu}$ ($\mu \sim 0.8$) of the collective-flux-creep model in the regime $J < J_{c0}$, while properly describing the finite pinning potential observed in the activated-flux-flow regime as $J \rightarrow 0$. The resulting analysis using this form for U_0 provides a quantitatively self-consistent interpretation of all sets of measurements.

The description of flux motion in high-T_c superconductors is an interesting and complicated problem. The relationship between dissipative flux motion and the flux-pinning mechanism has been discussed extensively, but is still not well understood. Because the effective pinning potential barrier $U(J,T,B)$ is an important parameter for interpreting the flux motion, several expressions have been proposed by various authors.[1–7] Moreover, since the thermal energy kT in the superconducting state of high-T_c oxides can be much higher than for conventional superconductors, the effect of thermally-activated flux motion is stronger, even for pinning energies of comparable magnitude. These effects have been considered[1,5] and the flux-motion state described in terms of thermally-activated processes.

Recently, Fisher indicated that there is a glasslike vortex phase at low magnetic field,[2] and experimental evidence was given to support a proposed vortex-melting transition at a characteristic temperature.[8] In the vortex-glass phase, flux motion is described by a power-law dependence of the effective pinning barrier on the current density J, with $U \propto J^{-\mu}$. The same dependence is derived from the theory of collective flux creep,[3,4] which depicts the hopping of coherent bundles of flux lines. Within vortex-glass theory, it is thought that the exponent μ has some universal value, while in the collective-flux-creep model μ varies with the magnetic field and depends on the effective dimensionality of the flux-line lattice (FLL). Experimentally, substantial evidence has accumulated for a field-[9] and temperature-dependent[7] exponent μ. This general formalism has also provided a good modeling of the quasiexponential temperature dependence of $J_c(T)$ at lower temperature,[10] as well as flux-creep–magnetic-relaxation studies[11] on $YBa_2Cu_3O_{7-\delta}$ (YBCO) samples with various oxygen stoichiometries.

Another model for $U(J)$ was proposed by Zeldov *et al.*,[6] who obtained $U \propto -\ln J$, based on observed resistive transitions of an epitaxial film as a function of current density J and magnetic field B. Subsequent determinations of $U(J)$ from magnetic-relaxation experiments on single crystals have indicated a similar form.[12]

These different J dependencies of U lead to different predictions for the transport characteristics as a function of magnetic field B and temperature T. Here we have studied the field- and temperature-dependent transport properties of a series of high-J_c epitaxial YBCO thin films. The characteristic flux motion in these thin films is consistent with a modified flux-creep model, signified by an exponential dependence of U on current density. This model is founded on related work that explored the flux-pinning scaling behavior in epitaxial YBCO films[13] and the long-term magnetic-flux relaxation of high-J_c, proton-irradiated YBCO single crystals.[7] It is shown that the current-voltage relationships below and above the irreversibility line are systematically consistent; furthermore, the experimental data in these respective regimes of creep-limited J_c and of dissipative flux flow can be described by the theory of thermally-activated flux motion.[5]

The YBCO c-axis-perpendicular epitaxial thin films were prepared on (001) surfaces of various substrates $LaAlO_3$, $SrTiO_3$, YSZ, and $KTaO_3$ either *in situ* by laser ablation[14] or *ex situ* by coevaporation and post-annealing.[15,16] The transport measurements on patterned films were carried out by a standard four-probe dc technique.[17] I-V curves were taken at various fixed temperatures for magnetic fields B parallel to the c axis, and the criterion of critical-current density J_c was set at 1 μV/cm. In-field resistive transitions were measured using transport-current densities in the range of a few A/cm^2.

In the standard flux-creep model, the flux-bundle hopping frequency is given by

$$\nu=\nu_0 \exp\left[-\frac{U_0}{kT}\right]\sinh\left[\frac{W}{kT}\right], \tag{1a}$$

$$W=JBV_bX_p\ . \tag{1b}$$

Here ν_0 is a characteristic attempt frequency and W is the work done by the Lorentz driving force against U_0, resulting in the effective pinning potential $U=U_0-W$, where U_0 is the J-dependent activation barrier of range X_p.[5] The flux-bundle volume V_b may be taken to be the one defined by Larkin and Ovchinnikov[18] in the context of collective processes. Note that we do not assume that U_0=const, as in the traditional Anderson-Kim formula. If a FLL bundle moves with velocity $\mathbf{v}$, an electric field $\mathbf{E}=\mathbf{B}\times\mathbf{v}/c$ is generated by the driving force, resulting in nonlinear $E(J)$ curves

$$E=E_0 \exp\left[-\frac{U_0}{kT}\right]\sinh\left[\frac{W}{kT}\right], \tag{2}$$

where $E_0=\nu_0 Bx/c$ and $U_0=U_0(J,T,B)$; x is an average hopping distance.

In the critical state and in the absence of thermal activation, the driving force resulting from the critical-current density J_{c0} is compensated by the pinning force such that

$$U_0(J_{c0},T,B)=W=J_{c0}BV_bX_p\ . \tag{3}$$

Depending on the details of the flux-pinning system, J_{c0} is some particular function of B and T. For example, from interpretation of the present experiments[13] and on theoretical grounds,[18] we have

$$J_{c0}(T,B)\simeq J_{c0}(T,0)\left[1+\frac{B}{B_0}\right]^{-n}(1-b)\ ,$$

where $b=B/B_{c2}$, $B_{c2}(T)$ is the upper critical field, $B_0(T)$ represents a field below which intervortex interactions become negligible, and $n\approx 1-1.5$.

We now make a plausibility argument for the observed dependence $U_0 \propto e^{-J/J_{c0}}$. In general, the jumping distances and FLL-bundle volumes vary with current, temperature, and magnetic field.[3,4,19,20] In addition, we consider a distribution of pinning potentials due to the various pinning strengths and mechanisms,[19] such that when an external current passes through the a-b planes, some FLL bundles remain pinned because the driving force is not strong enough to overcome the total pinning force. From Eq. (1) the work W increases with J while U_0 decreases as a result of the reduction[3] in volume V_b. Because of the conditions that $W=0$ at $J=0$ and that $W=U_0(J_{c0})$ at $J=J_{c0}$, we propose the following relation between an average U_0 and W in terms of the reduced current density $j=J/J_{c0}$:

$$W(j,T,B)\simeq U_0(j,T,B)+(1-j)\frac{dU_0(j,T,B)}{dj}\ . \tag{4}$$

The second term comes from that part of the activation energy which is larger than the driving force. The present experimental data and previous observations[1,21,22] show that $W\propto j$ when $j\ll 1$, and from Eqs. (1) and (3) it is seen that at large j, $W\simeq jU_0$. Substitution in Eq. (4) yields a solution

$$U_0(j,T,B)=U_0(T,B)\exp(-j)\ . \tag{5}$$

Then Eqs. (2) and (5) lead to the general result

$$E=E_0 \exp\left[-\frac{U_0(T,B)}{kT}\exp(-j)\right]\times\sinh\left[\frac{jU_0(T,B)}{kT}\exp(-j)\right]. \tag{6}$$

In the following we employ Eq. (6) to describe the transport properties over a range of current densities, fields, and temperatures, both above and below the irreversibility line $B_{\rm irr}(T)$.

First, we use this result to interpret the transport properties in the dissipative regime for $B>B_{\rm irr}$, where Eq. (6) yields, in the limit of small j,

$$\rho=\frac{E}{J}=\frac{E_0(T,B)}{J_{c0}(T,B)}\frac{U_0(T,B)}{kT}\left[1+j\left[\frac{U_0(T,B)}{kT}-1\right]\right]\times\exp\left[-\frac{U_0(T,B)}{kT}\right]. \tag{7}$$

This flux-hopping resistivity corresponds to the linear portion of $E(J\to 0)$. Data for several epitaxial films were analyzed, and the results are typified by the plots in Fig. 1, which pertains to a film deposited on $LaAlO_3$. The Arrhenius-like plots of $\ln(\rho)$ vs $1/B$ are shown for several temperatures in the range $T>77$ K, where the region $B>B_{\rm irr}$ is experimentally accessible. In the limit of vanishing j, the slope of these plots gives directly the quantity U_0B/kT. Over several decades in ρ, the $\ln(\rho)$-vs-$1/B$ curves are substantially linear, indicating that $U_0\propto 1/B$, except perhaps very close to the normal-state boundary where inhomogeneity and fluctuation effects may become

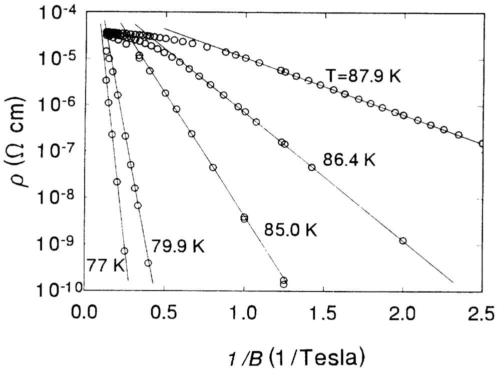

FIG. 1. Arrhenius-type plot of resistivity ρ vs $1/B$ for $B > B_{irr}$, at the fixed temperatures shown. Solid lines are fitted to Eq. (7) with $j \to 0$.

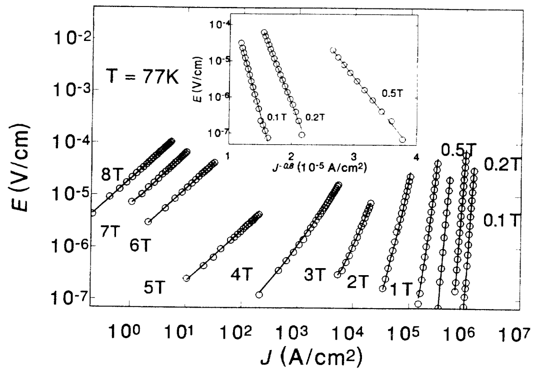

FIG. 3. Measured $E(J)$ at 77 K for a YBCO film deposited on $LaAlO_3$, for several applied magnetic fields. The solid curves are fits to Eq. (6), while the inset shows that, at low fields, the data are also consistent with the predictions of vortex-glass–collective-flux-creep models, where $E \propto \exp(-j^{-\mu})$, with $\mu \approx 0.8$.

important. With this observed field dependence extracted, we illustrate in Fig. 2 the derived temperature dependence of the pinning energy. By plotting the product $U_0 B$ vs a standard, high-temperature thermal factor $(1-T/T_c)$, a power-law dependence is found, with exponent ≈ 1.8 Incorporating these dependencies, we conclude that, in this high-temperature range, $U_0(j,T,B) = U_{00} e^{-j}(1-b)/b(1-t)^{1.8}$, with $U_{00} \approx 6800$ K = 0.58 eV. For the other films investigated, U_{00} values fall in the range of 5000–20 000 K. For completeness, we have included here the factor $(1-b)$, which must be present to account for overall gap suppression by the average magnetic field, but the factor is unimportant in the range of fields involved in the linear analyses.

We now show that Eq. (6) also is quantitatively consistent with the transport properties below the irreversibility line. Figure 3 is a set of E-J curves taken at $T=77$ K for magnetic fields from 0.1 to 8 T. For $B < 1$ T, the curves are nearly linear with slight negative curvature, while those curves taken above 1T have positive curvature, as indicated in Eq. (7). The solid curves are fits to Eq. (6), where $U_0(T,B)$ is a deduced parameter to be discussed below and values for J_{c0} were taken from an analysis of quasiscaling of the pinning-force density[13] on the same sample. Interestingly, at low fields, where $j < 1$, the main term of Eq. (6) mimics that given by a power-law current dependence of the pinning energy, $U \propto j^{-\mu}$. This fact is illustrated in the inset, which shows that the present data can be fit by the vortex-glass–collective-creep model with $\mu \approx 0.8$. This latter value is consistent with Fisher's limit[2] that $\mu \leq 1$; however, the exponent μ decreases[13] rapidly for $B > 1$ T. This observation is qualitatively similar to the decrease with field that was observed in flux-creep studies[9] on proton-irradiated single crystals of YBCO.

Results for $U_0(T,B)$, obtained from fits to the shape of the $E(J)$ curves as described by Eq. (6), are presented in Fig. 4 vs $1/B$. For $B > 1$ T, the previously deduced dependence $U_0(T,B) \propto 1/B$, derived from the activated resistance for $B > B_{irr}$, is again observed both above and below B_{irr} as illustrated in the inset. At low fields $B < B_0$, U_0 approaches saturation at a final value corresponding to single-vortex pinning in the dilute limit.

In Fig. 5 experimental temperature-dependent resistive

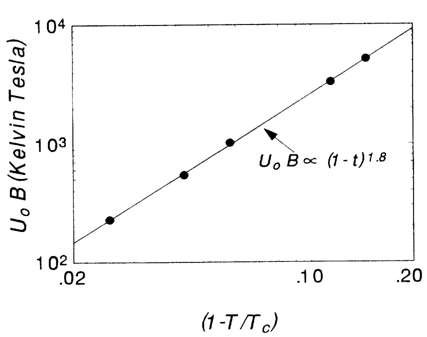

FIG. 2. Temperature-dependent component of the activation barrier U_0, plotted as the product $U_0 B$. A linear regression (solid line) gives $U_0(T) \propto (1-t)^{1.8}$ in this regime, using the observed $T_c(R=0)=90$ K.

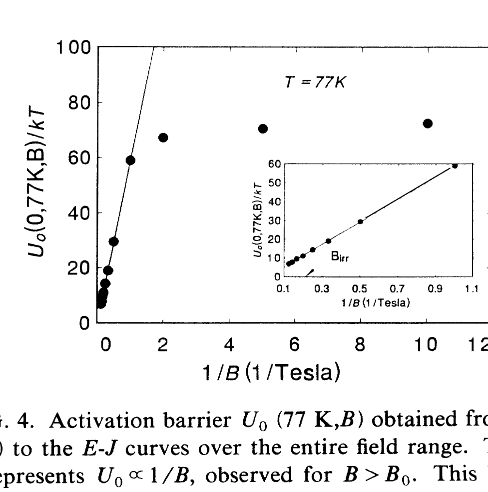

FIG. 4. Activation barrier U_0 (77 K,B) obtained from fits of Eq. (6) to the E-J curves over the entire field range. The solid line represents $U_0 \propto 1/B$, observed for $B > B_0$. This latter regime is shown on an expanded scale in the inset. Note that $U_0 \propto 1/B$ for both $B > B_{irr}$ and $B < B_{irr}$.

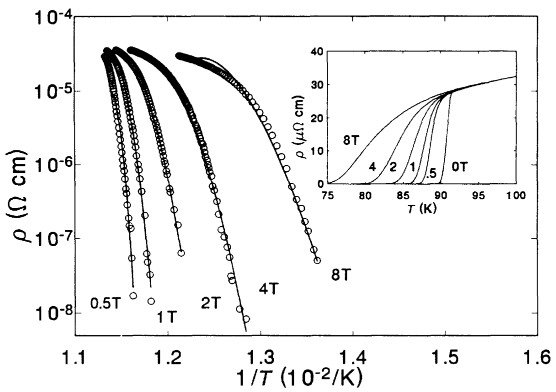

FIG. 5. Arrhenius plots of the resistive transition in the magnetic fields indicated. Solid curves were generated from Eq. (7), using the parameters previously determined from an analysis of E-vs-J measurements (Fig. 4). The model deviates at temperatures where full flux-flow and fluctuation effects become prominent.

transitions in a magnetic field for the same YBCO film are plotted on an Arrhenius diagram. The solid curves were generated by substituting into Eq. (7) the parameters obtained in the analysis of Fig. 4; a temperature dependence of the form $(1-t)^m$ was used, with the m to be determined experimentally. The results have only logarithmic sensitivity to the temperature dependencies of $E_0(T,B)$ and $J_{c0}(T,B)$, but are strongly influenced by the B and T dependencies of $U_0(T,B)$. The agreement is good in the extensive low-ρ region, but fails as Eq. (7) loses validity, when flux-flow and perhaps fluctuation effects dominate close to the resistive onset at $T_c(B)$. From Fig. 5 we obtain a temperature dependence $U_0(T) \propto (1-t)^m$, with $m = 1.8 \pm 0.1$, which agrees well with both theoretical predictions[23] and the experimental observations of Fig. 2. It is worthwhile to emphasize that the slope of the curves in Fig. 5, at any field B and temperature T, yields the local value of $U_0(B,T,j \to 0)/k$. It can be seen that curvature persists over a wide range of T, pointing up the difficulty in extracting a representative value of the temperature-dependent U_0 from a single linear fit to $\ln(\rho)$ vs $1/T$.

In summary, we have shown that the high-temperature superconductive transport properties of high-J_c epitaxial thin films can be quantitatively and self-consistently described in terms of a model of thermally-activated flux motion, with a pinning activation barrier that has the form $U_0 \propto (1-T/T_c)^{1.8} \exp(-J/J_{c0})/B$. Within the experimental detection limit and at the magnetic fields and temperatures accessible, we find that this form duplicates the results of the vortex-glass and collective-pinning theory in the regime $J_c < J_{c0}$ [i.e., well below the irreversibility line $B_{\mathrm{irr}}(T)$]. Moreover, this potential properly accounts for the intermediate cases $J_c < J_{c0}$ and successfully describes the finite pinning potential observed in the activated-flux-flow regime $B > B_{\mathrm{irr}}$ as $J \to 0$. The J dependence of U_0 discussed here applies as well to other work[13] on the observed quasiscaling of the pinning-force density F_p with respect to a scaling field $B^* \approx B_{\mathrm{irr}}$.

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[1]Y. Yeshurun and A. P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).

[2]M. P. A. Fisher, Phys. Rev. Lett. **62**, 1415 (1989).

[3]M. V. Feigelman, V. B. Geshkenbein, A. I. Larkin, and V. M. Vinokur, Phys. Rev. Lett. **63**, 2303 (1989).

[4]T. Natterman, Phys. Rev. Lett. **64**, 2454 (1990).

[5]P. H. Kes, J. Aarts, J. van ven Berg, C. J. van der Beek, and J. A. Mydosh, Supercond. Sci. Technol. **1**, 242 (1989).

[6]E. Zeldov, N. M. Amer, G. Koren, A. Gupta, M. W. McElfresh, and R. J. Gambino, Appl. Phys. Lett. **56**, 680 (1990).

[7]J. R. Thompson, Y. Sun, and F. H. Holtzberg, Phys. Rev. B **44**, 469 (1991).

[8]R. H. Koch, V. Foglietti, W. G. Gallagher, G. Koren, A. Gupta, and M. P. A. Fisher, Phys. Rev. Lett. **63**, 1511 (1989).

[9]J. R. Thompson, Y. R. Sun, D. K. Christen, H. R. Kerchner, A. P. Malozemoff, L. Civale, A. D. Marwick, L. Krusin-Elbaum, M. W. McElfresh, and F. Holtzberg, in *Physics and Materials Science of High Temperature Superconductors-II*, Vol. 209 of *NATO Advanced Study Institute, Series E: Applied Sciences*, edited by R. Kossowsky, B. Raveau, D. Wohlleben, and S. Patapis (Kluwer Academic, Dordrecht, in press).

[10]J. R. Thompson, Y. R. Sun, L. Civale, A. P. Malozemoff, M. W. McElfresh, A. D. Marwick, and F. Holtzberg (unpublished).

[11]J. G. Ossandon, J. R. Thompson, D. K. Christen, B. C. Sales, Y. R. Sun, and K. W. Lay, Phys. Rev. B **46**, 3050 (1992).

[12]M. P. Maley, J. O. Willis, H. Lessure, and M. E. McHenry, Phys. Rev. B **42**, 2639 (1990).

[13]D. K. Christen, S. Zhu, C. E. Klabunde, R. Feenstra, D. H. Lowndes, and D. P. Norton (unpublished).

[14]Douglas H. Lowndes, David P. Norton, J. W. McCamy, R. Feenstra, J. D. Budai, D. K. Christen, and D. B. Poker, in *High Temperature Superconductors: Fundamental Properties and Novel Materials Processing,* edited by D. Christen, J. Narayan, and L. Schneemeyer (Materials Research Society, Pittsburgh, 1990), Vol. 169, p. 431.

[15]R. Feenstra, L. A. Boatner, J. D. Budai, D. K. Christen, M. D. Galloway, and D. B. Poker, Appl. Phys. Lett. **54**, 1063 (1989).

[16]R. Feenstra, T. B. Lindemer, J. D. Budai, and M. D. Galloway, J. Appl. Phys. **69**, 6569 (1991).

[17]D. K. Christen, C. E. Klabunde, R. Feenstra, D. H. Lowndes, D. Norton, H. R. Kerchner, J. R. Thompson, S. T. Sekula, J. D. Budai, L. A. Boatner, J. Narayan, and R. Singh, in *High*

Temperature Superconductors: Fundamental Properties and Novel Materials Processing (Ref. 14), p. 883.
[18]A. I. Larkin and Yu. N. Ovchinnikov, J. Low Temp. Phys. **34**, 409 (1979).
[19]A. Gurevich, Phys. Rev. B **42**, 14 857 (1991).
[20]K. H. Fischer and T. Nattermann, Phys. Rev. B **43** 10 372 (1991).
[21]T. T. M. Palstra, B. Batlogg, R. B. van Dover, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. B **41**, 6621 (1990).
[22]J. D. Hettinger, A. G. Swanson, W. J. Skocpol, J. S. Brooks, J. M. Graybeal, P. M. Mankiewich, R. E. Howard, B. L. Straughn, and E. G. Burkhardt, Phys. Rev. Lett. **62**, 2044 (1989).
[23]M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988).