Scaling of the Flux Pinning Force in Epitaxial $Bi_2Sr_2Ca_2Cu_3O_x$ Thin Films

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Magnetic-field and temperature dependence of the critical current density J_c is investigated in epitaxial $Bi_2Sr_2Ca_2Cu_3O_x$ thin films. For the magnetic field H applied parallel to the c axis, the flux pinning force density F_p ($=J_cB$) exhibits clear scaling behavior when H is normalized by the irreversibility field H^*. The maximum pinning force density scales linearly with H^*. This is the first observed scaling of F_p in high-quality thin films of Bi oxides, which we can reasonably explain with flux-creep theory by assuming that the activation energy is proportional to the flux line spacing.

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Bi oxide cuprate superconductors, $Bi_2Sr_2CaCu_2O_x$ (Bi-2212) and $Bi_2Sr_2Ca_2Cu_3O_x$ (Bi-2223), are technologically important because relatively high transport critical current densities, $J_c \sim 10^8-10^9$ A/m^2, can be obtained in polycrystalline samples such as Ag-sheathed tapes [1,2]. J_c, however, declines precipitously with magnetic field at moderate temperatures (> 30 K) because of weak flux pinning [1]. The mechanism controlling J_c in the Bi oxides is one of the key issues for the application of this material. The irreversibility temperatures and the irreversibility fields H^* above which magnetization becomes reversible and hence J_c becomes zero are much lower than those observed in $YBa_2Cu_3O_7$ (YBCO) [3].

Recently, we prepared high-quality, epitaxial Bi-2223 thin films by metalorganic chemical vapor deposition, showing high critical temperatures T_c of 92–97 K [4]. These films have the highest reported J_c for the Bi-oxide system: $J_c \gtrsim 10^9$ A/m^2 at 77.3 K in high magnetic fields (1–8 T, $H \| a$-b plane) [4,5]; zero field $J_c = 1.3\times10^{10}$ A/m^2 at 70 K and 10^{11} A/m^2 at 30 K [6]. In this Letter we report clear scaling behavior over a wide temperature range, 20–60 K, for the flux pinning force density F_p, calculated from J_c values measured in fields parallel to the c axis. Such F_p scaling has been observed in the transport J_c of YBCO polycrystalline thin films [7] and epitaxial films [8,9] in limited temperature ranges, but until now was not reported for high-quality Bi-oxide thin films. The F_p scaling, which is technologically important in predicting the J_c behavior in magnetic fields, is reasonably explained using flux-creep theory by assuming that the activation energy is proportional to the flux line spacing a_0.

The details of sample preparation have been reported elsewhere [4]. The 60–80 nm thick films used in this study, deposited on $LaAlO_3$ (100) single-crystal substrates, were characterized by x-ray diffraction to be single-phase Bi-2223 thin films with the c axis oriented perpendicular to the film surface. High-resolution scanning electron microscopy and reflection high-energy electron diffraction observations confirmed that they were epitaxially grown without apparent grain boundaries.

The transport J_c was measured by a four-probe dc method for bridge patterns with narrow strip lines, 2 mm long and 50 μm wide, made by a chemical etching technique. Evaporated silver film pads were used as electrodes, which were annealed at 300 °C to reduce the contact resistivity. J_c is defined by a criterion of 2 μV/cm electric field. Magnetic fields were always applied perpendicular to the transport current.

$J_{c\|}(H,T)$ measured in applied fields parallel to the film surface (a-b plane) was almost field independent at temperatures $\lesssim 60$ K [Fig. 1(a)], while $J_{c\perp}(H,T)$ measured in fields perpendicular to the a-b plane decreased sharply with field except at low temperatures (≤ 20 K) [Fig. 1(b)]. Such anisotropic J_c behavior comes from the quasi-two-dimensional nature of the Bi oxides [6,10,11].

For magnetic fields applied parallel to the c direction, the flux pinning force density $F_p = J_{c\perp}(H,T)B$ was found to scale with the irreversibility field H^* and the maximum pinning force density $F_{p_{max}}$. H^* was determined to

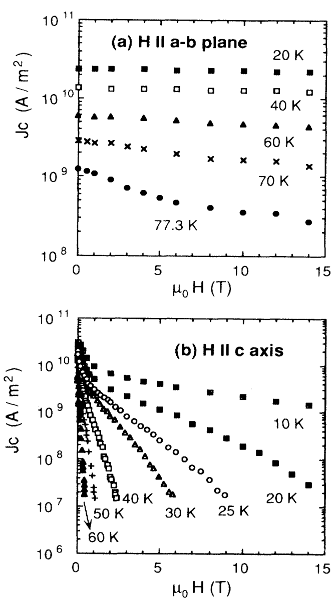

FIG. 1. Critical current density J_c measured in a Bi-2223 thin film at various temperatures (a) in magnetic fields applied parallel to the a-b plane $J_{c\|}(H,T)$ and (b) in fields parallel to the c axis $J_{c\perp}(H,T)$.

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be the field at which J_c defined in this particular case by the criterion of 1 μV/cm is less than 10^7 A/m^2 [5]. The H^* values are not dependent on the definition much because of the steep J_c decline with magnetic field at $T \geq 30$ K [12]. When the magnetic field was normalized as $h = H/H^*$, clear scaling behavior was observed in F_p (Fig. 2). Similar scaling was also observed in the other two samples with lower J_c's over the temperature range 20–60 K. The $F_{p_{\max}}$ values scale linearly with H^* for three different Bi-2223 thin film samples as depicted in the inset of Fig. 2. The physical interpretation of these data will be given later. For YBCO a scaling of the form $F_p = K(H^*)^n\sqrt{h}(1-h)^2$ has been reported for a limited temperature range (71–81 K) [7,9]. However, too large H^* values at lower temperatures hindered the observation of the exact scaling behavior over a wider temperature range [7,8], and no analysis has been able to explain the F_p scaling for YBCO satisfactorily.

Figure 3 shows the Arrhenius plots of the resistivity $\rho(H,T)$ measured in fields parallel to the c axis for the same sample whose J_c data are shown in Fig. 1. $T_c(R=0)$ was $\sim$86.5 K. Except near $1/T_c$ where superconducting fluctuations set in, these plots are almost linear in a wide $1/T$ range, which indicates that the resistance is caused by a thermally activated process. Temperature independent linear logρ-$1/T$ plots have been reported for Bi-2212 single crystals [13] and thin films [14]. From the data of Fig. 3 the activation energy U_0 was calculated and plotted against H in the inset of Fig. 3. The $\rho(H,T)$ data were taken with small $J = 2.5\times10^6$ A/m^2. Essentially the same ρ-T curve was also obtained using a smaller current density, which means that the resistivity has been measured in the linear portion of the J-E characteristic. In this experiment, therefore, the calculated U_0 is for the limit $J \approx 0$. The inset of Fig. 3 indicates that U_0 is proportional to $H^{-0.51} \approx 1/\sqrt{H}$. This $1/\sqrt{H}$ dependence of U_0 has been demonstrated in the detailed analyses of $\rho(H,T)$ data in Bi-2212 thin films [14,15] and has also been observed in Bi-2212 wires [16], although a $1/H$ dependence has been observed in YBCO [8,17].

The physical origin of the observed $1/\sqrt{H}$ dependence of U_0 is proposed by Geshkenbein *et al.* [18]. In this model U_0 is associated with the plastic deformation of flux line lattice (FLL) at FLL dislocations, analogous to the thermally activated motion of edge dislocations in crystals. U_0 is estimated as the energy required to create a double kink in the flux line,

$$U_0 = 2\varepsilon_1 a_0 \approx (\Phi_0^2/2\pi\mu_0\lambda^2)\ln\kappa(\Phi_0/B)^{1/2}$$
$$= \mu_0 H_c^2 4\pi\xi^2 \ln\kappa(\Phi_0/B)^{1/2},$$

where ε_1 is the extra energy per unit length of the flux line along the CuO_2 plane, λ the penetration depth, ξ the coherence length, and $\kappa = \lambda/\xi$ [18]. This model is similar to Tinkham's model of thermally activated vortex lattice shear which explains the $1/H$ dependence for YBCO [19], in that "the motion of a row of fluxons past neighbor rows" [18] occurs. However, in highly anisotropic Bi-oxide system, FLL deforms plastically forming double kinks rather than shearing without kinking [14,18].

We discuss the F_p scaling observed in our Bi-2223 thin films based on the theory of flux creep [20–22]. In intermediate magnetic-field range, $H_{c_1} \ll H \ll H_{c_2}$, collective effects of flux lines are important. The electric field induced by the motion of "flux bundles" is given by $E = Bd\nu_0\exp(-U_{\rm eff}/kT)$, where d is a distance which the flux bundles move (of the order of a_0), ν_0 is an attempt frequency, and $U_{\rm eff}$ is an effective activation energy. Thermally activated flux motion is assisted by the driving force $F = JB$, and in the original Anderson-Kim model $U_{\rm eff}$ is expressed as $U_{\rm eff} = U_0 - JBV_aX_p$, where V_a is the activation volume and X_p is the effective range of the potential well [20]. If $J_0 = U_0/BV_aX$ is defined as the zero-temperature pinning force density when there is no thermal activation and $j = J/J_0$, $U_{\rm eff}$ can be written as $U_{\rm eff} = U_0(1-j)$.

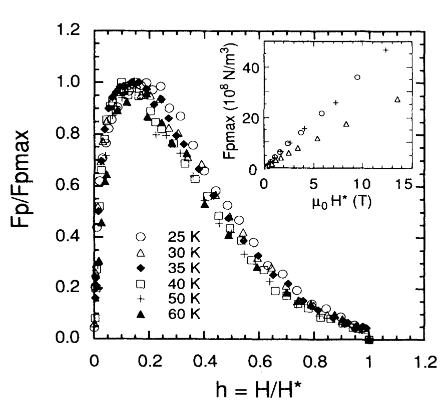

FIG. 2. Scaling of the flux pinning force density $F_p = J_cB$ measured with the field parallel to the c axis. Inset: Relationship between $F_{p_{\max}}$ and H^* for three different thin film samples.

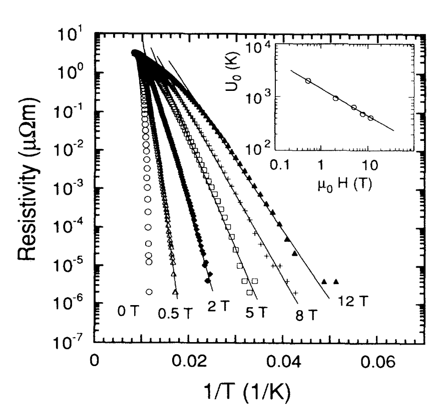

FIG. 3. Arrhenius plots of the resistivity $\rho(H,T)$ for the field parallel to the c axis. Inset: Activation energy U_0 plotted against applied field H.

Until now there have been many reports that suggest that U_{eff} is not a linear function of J, but has an upward curvature [17,23–25], which was already pointed out in the classical paper of Beasley, Labusch, and Webb [21]. Recent study on YBCO epitaxial films observed an E-J relation explained by $U_{eff}=U_0(1-j)\exp(-j)$ [17]. In our present study we assume a sinusoidal potential $U(X) = \frac{1}{2}U_0\cos(\pi X/X_p) - JBV_aX$ and $U_{eff}=U_0(1-j)^{3/2}$ [21, 24], which has a j dependence comparable to $U_0(1-j)\times\exp(-j)$. It is to be noted that the functional form of $U_{eff}(j)$ does not much affect our analysis: The classical Anderson-Kim model was able to give a result similar to the present study. $U_{eff}=U_0(1-j)^{3/2}$ leads to

$$E=Bdv_0\exp[-U_0(1-J/J_0)^{3/2}/kT]\,. \quad (1)$$

In our transport J_c measurement J_c was defined by an electric field criterion $E_c=2\ \mu$V/cm. By setting $E=E_c$ and $J=J_c$ in Eq. (1),

$$J_c=J_0\{1-[(kT/U_0)\ln(Bdv_0/E_c)]^{2/3}\}\,. \quad (2)$$

Since $\ln(Bdv_0/E_c)$ is a slowly varying function of H, we set $\ln(Bdv_0/E_c)=\ln(E_0/E_c)$ to be a constant [26]. When U_0 is expressed as $U_0=\alpha(T)/\sqrt{H}$, as observed in our $\rho(H,T)$ data and in Ref. [14], and H_0 is defined as $(H_0)^{1/2}=\alpha(T)/[kT\ln(E_0/E_c)]$, $F_p=J_cB$ is calculated to be

$$F_p=J_0B[1-(H/H_0)^{1/3}]\,. \quad (3)$$

We next need to calculate $J_0B=\pi U_0/2V_aX_p$. Since there have been no experimental results from which V_a and X_p can be estimated, we will make the following speculations. If the Lorentz force is exerted effectively on a row of flux lines having number N, the activation volume V_a can be expressed as $Na_0\xi_{ab}d_k$, where d_k is the length of the flux line along the c axis between the two kinks formed. If we assume that the flux lines are weakly pinned by prevalent pinning centers like point defects, the movable length d_k can be proportional to kT. The potential well range X_p is approximated by $a_0\approx(\Phi_0/B)^{1/2}$. Since ξ_{ab} is weakly temperature dependent for low t $(=T/T_c)$ values,

$$J_0B\approx\mu_0K\alpha(T)\sqrt{H}/kT=\mu_0K\sqrt{H_0}\sqrt{H^*}\sqrt{h}\ln(E_0/E_c)\,, \quad (4)$$

where $K=\pi[2N\xi_{ab}\Phi_0(d_k/kT)]^{-1}$. Putting $H^*=\beta^2H_0$, we finally obtain

$$F_p=(\mu_0K/\beta)H^*\ln(E_0/E_c)\sqrt{h}(1-\beta^{2/3}h^{1/3})\,, \quad (5)$$

which can explain the general feature of the F_p scaling and the linear H^*-$F_{p_{max}}$ relationship shown in Fig. 2. The curve fitting to the F_p data of 30 and 40 K in the range of $0<h<0.4$ with $\beta=1.25$ is shown in Fig. 4. Here, the intersection of the curve with the horizontal axis gives H_0. We point out that in Eq. (4) J_0B is a monotonically increasing function of h (and H). This seems to be contradictory to the F_p behavior observed for conventional low-T_c superconductors [27,28]: The field dependence of F_p can be expressed as $(H/H_{c2})^p(1-H/H_{c2})^q$. Since the upper critical fields of Bi oxides are very high compared with H^*, e.g., $\mu_0H_{c2}\approx50$ T at 60 K for Bi-2212 single crystals [29], the term $1-H/H_{c2}$, which describes the decrease of the mean superconducting condensation energy with the magnetic field [30], can be neglected in our analysis.

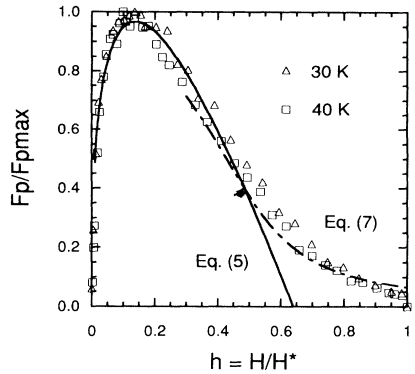

FIG. 4. Curve fitting by Eqs. (5) and (7) to the F_p data of 30 and 40 K.

Up to the present stage we have neglected the possibility of flux motion in the opposite direction toward a higher energy barrier by assuming that $\exp(JBV_aX_p/KT) \gg\exp(-JBV_aX_p/kT)$. However, this is not the case when the driving force gradient is small compared with kT. The equation which takes into account the probability of flux motion in both directions is [22,24]

$$E=2E_0\exp\{-U_0[(1-j)^{3/2}+(\pi/2)j]/kT\}\times\sinh(\pi U_0j/2kT)\,. \quad (6)$$

This equation is applied for large h $(0\ll h<1)$ where $j\ll1$, and is reduced to $E=2E_0\exp(-U_0/kT)\sinh(\pi U_0j/2kT)$, because $(1-j)^{3/2}+(\pi/2)j\approx1$. By following the same procedure used to obtain Eq. (5), we get

$$F_p=(2/\pi)\mu_0KH^*h\times\sinh^{-1}(\tfrac{1}{2}\exp\{[(1/\beta\sqrt{h})-1]\ln(E_0/E_c)\})\,. \quad (7)$$

The curve fitting using Eq. (7) with $\ln(E_0E_c)=8$ is also shown in Fig. 4. The experimental $F_p/F_{p_{max}}$ data agree well with the theoretical result. Equations (5) and (7) have three independent parameters—K, β, and $\ln(E_0/E_c)$. β and $\ln(E_0/E_c)$ were determined to be 1.25 and 8, respectively, by the curve fitting shown in Fig. 4. Then K was calculated to be 3.8×10^8 A/m^2 from the linear $F_{p_{max}}$-H^* relationship shown in the inset of Fig. 2.

The above analysis based on flux-creep theory explains why F_p scales with $h=H/H^*$, not with H/H_{c2}. Similar treatment was done by Hettinger *et al.* [8] to explain the F_p scaling observed in YBCO thin films. They overlooked, however, the upward curvature of the U-J relation (inset of Fig. 1 of Ref. [8]), which leads to an underestimation of J_0, a lower J_c than observed at 20 K, and an overestimation of V_d, the volume over which the driving force is determined. If V_d has the same field dependence as V_a in Ref. [8], a similar treatment to that used to obtain Eq. (5) in this study leads to $F_p \sim \sqrt{h}(1-\beta h^{2/3})$ in low h regime, which better explains their F_p data. They assumed the individually pinned vortices regime to explain the increase of F_p in low h regime ($h \leq 0.15$). In this regime, however, field independent J_c is expected and the regime threshold is not necessarily proportional to H^* [31], which is contradictory to their experimental results and interpretation. Civale *et al.* assumed a different form $J_0 \propto 1/[1+B/B_0(T)]$ in their analysis on the magnetization scaling in YBCO single crystals [32].

The major assumption in our analysis is that the $1/\sqrt{H}$ dependence of U_0 derived from the relation $U_0 \propto a_0$, which has been generally observed in Bi-oxide systems for ρ-T data taken above the irreversibility line (IL), is also the case in the discussion of J_c below IL. For the highly anisotropic Bi system the theory of Geshkenbein *et al.* [18] explains the temperature and field dependence of low U_0 values at high temperatures and low current densities. It is reasonable to think that their theory can also be applicable at lower temperatures and high current densities, since 2D-like J_c behavior was observed also at low temperatures. The stepwise flux penetration with many kinks is expected because the blocking layers between the superconducting CuO_2 layers have a lower superconducting order parameter [10], which is confirmed by the angular dependence of $J_c(H,\theta)$ determined only by the magnetic field component along the c direction, $J_c(H,\theta) = J_{c\perp}(H\sin\theta)$ [6,11]. This suggests that the flux motion with formation of double kinks is plausible also below IL. In recent transport measurements in epitaxial YBCO films the $1/B$ dependence of U_0 is observed *both above and below* the irreversibility field [17], which gives a justification to our analysis.

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[1] M. P. Maley, J. Appl. Phys. **70**, 6189 (1991).
[2] K. Sato *et al.*, IEEE Trans. Magn. **27**, 1231 (1991).
[3] Y. Yeshurun *et al.*, Cryogenics **29**, 258 (1989).
[4] K. Endo *et al.*, Nature (London) **355**, 327 (1992).
[5] H. Yamasaki *et al.*, J. Appl. Phys. **72**, 2951 (1992).
[6] H. Yamasaki *et al.*, IEEE Trans. Appl. Supercond. (to be published).
[7] J. S. Satchell *et al.*, Nature (London) **334**, 331 (1988).
[8] J. D. Hettinger *et al.*, Phys. Rev. Lett. **62**, 2044 (1989).
[9] T. Nishizaki *et al.*, Physica (Amsterdam) **181C**, 223 (1991).
[10] M. Tachiki and S. Takahashi, Solid State Commun. **70**, 291 (1989); **72**, 1083 (1989).
[11] P. Schmitt, P. Kummeth, L. Schultz, and G. Saemann-Ischenko, Phys. Rev. Lett. **67**, 267 (1991).
[12] The irreversibility field H^* defined here is not the same as that determined by the magnetization measurement usually reported, because of different electric field critera. However, Xu and Suenaga reported that resistively defined H^* values have a temperature dependence comparable to magnetically determined H^* values [Y. Xu and M. Suenaga, Phys. Rev. B **43**, 5516 (1991)].
[13] T. T. M. Palstra, B. Batlogg, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. **61**, 1662 (1988).
[14] J. T. Kucera *et al.*, Phys. Rev. B **46**, 11 004 (1992).
[15] The apparent temperature independent linear $\log\rho$-$1/T$ plots can be explained by the linear temperature dependence of U_0, $U_0 = K(1-T/T_c)/\sqrt{H}$, which was observed in Bi-2212 thin films in low fields ($\mu_0 H \leq 1$ T) [14]. The $1/\sqrt{H}$ dependence of the activation energy at a constant temperature was confirmed in Ref. [14].
[16] J. Löhle *et al.*, J. Appl. Phys. **72**, 1030 (1992).
[17] S. Zhu *et al.*, Phys. Rev. B **46**, 5576 (1992).
[18] V. Geshkenbein *et al.*, Physica (Amsterdam) **162-164C**, 239 (1989); see also a review in Ref. [14].
[19] M. Tinkham, Phys. Rev. Lett. **61**, 1658 (1988).
[20] P. W. Anderson, Phys. Rev. Lett. **9**, 309 (1962); P. W. Anderson and Y. B. Kim, Rev. Mod. Phys. **36**, 39 (1964).
[21] M. R. Beasley, R. Labusch, and W. W. Webb, Phys. Rev. **181**, 682 (1969).
[22] M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975), p. 175.
[23] D. O. Welch *et al.*, *Advances in Superconductivity II* (Springer-Verlag, Tokyo, 1990), p. 655.
[24] R. Griessen, Physica (Amsterdam) **172C**, 441 (1991).
[25] C. J. van der Beek *et al.*, Physica (Amsterdam) **195C**, 307 (1992).
[26] When $B = 1$ T, $d \approx 50$ nm, and v_0 is estimated to be 10^5–10^{11} sec^{-1} [22], which gives $25 \leq Bdv_0/E_c \leq 2.5 \times 10^7$. Since $Bdv_0/E_c \gg 1$ and d may be proportional to $a_0 \approx (\Phi_0/B)^{1/2}$, then $\ln(Bdv_0/E_c)$ is a slowly varying function of H.
[27] E. J. Kramer, J. Appl. Phys. **44**, 1360 (1973).
[28] D. Dew-Hughes, Philos. Mag. B **55**, 459 (1987).
[29] L. Zhang, J. Z. Liu, and R. N. Shelton, Phys. Rev. B **45**, 4978 (1992).
[30] H. Ullmaier, *Irreversible Properties of Type II Superconductors* (Springer-Verlag, Berlin, 1975), p. 146.
[31] L. Krusin-Elbaum, L. Civale, V. M. Vinokur, and F. Holtzberg, Phys. Rev. Lett. **69**, 2280 (1992).
[32] L. Civale *et al.*, Phys. Rev. B **43**, 13 732 (1991).