Thermally activated dissipation and irreversibility fields of bismuth oxide superconductors

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Thermally activated dissipation is investigated in epitaxial Bi-2:2:2:3 films in magnetic fields parallel to the c axis. For a wide temperature T and field B range the resistivity ρ shows clear Arrhenius behavior with an activation energy $U_0=A(1-T/T_c)/\sqrt{B}$: $\rho(B,T)=\rho_0\exp(-U_0/kT)=\rho_0\exp[-A(1-T/T_c)/kT\sqrt{B}]$, which is explained by thermally activated flux motion with the formation of double kinks. Temperature dependence of the irreversibility field B^* is predicted by flux-creep theory as $\sqrt{B^*}=C(1/kT-1/kT_c)$, when B^* is defined by an electric-field criterion. This relation holds not only for our films but also for magnetically determined B^* of Bi-2:2:2:3 samples, which suggests that the irreversibility line originates from thermally activated plastic motion of pinned flux liquid.

I. INTRODUCTION

The irreversibility field of type-II superconductors is defined as the field above which the magnetization becomes reversible and hence the critical current density J_c vanishes, as was first observed in $(La,Ba)_2CuO_4$ by Müller, Takashige, and Bednorz.[1] It is well known that the temperature dependence of the irreversibility field B^* of $YBa_2Cu_3O_7$ (YBCO) and $(La,Ba)_2CuO_4$ can be generally expressed by the power law $B^*\propto(1-T/T_c)^n$ with $n\approx1.5$, where T_c is the critical temperature.[1-3] This temperature dependence of B^* is attributed to the thermally activated flux motion or phase slip,[2,4] but the vortex-glass–liquid phase-transition theory has derived a similar relation $B_G\sim(T_c-T_G)^{4/3}$, where B_G and T_G are the glass-liquid transition field and temperature, respectively.[5-7] B^* values of Bi oxide superconductors, however, are much lower than those observed in YBCO,[8] and clear relationship between B^* and T has not been found.[3,8-12] A power-law function similar to that of YBCO can be applied to the low-field B^* data,[9-12] but B^* is better approximated by the exponential function $B^*=B_0\exp(-T/T_0)$ in higher fields.[9,10]

The irreversibility field B^* is a very important measure of the pinning force strength: a shift of the irreversibility line to higher fields and higher temperatures due to improved flux pinning is observed in irradiated YBCO (Ref. 13) and $Bi_2Sr_2CaCu_2O_x$ (Bi-2:2:1:2) (Ref. 14) single crystals, and the flux pinning force density $F_p=J_c\times B$ is scaled by B^* both in YBCO (Refs. 15–17) and in Bi oxides.[17,18] A detailed investigation of B^* is necessary to clarify the J_c behavior of oxide superconductors.

Recently we have prepared high-quality, epitaxial $Bi_2Sr_2Ca_2Cu_3O_x$ (Bi-2:2:2:3) thin films showing high-T_c values of 92–97 K.[19] These films, prepared by metal-organic chemical vapor deposition (MOCVD) on $LaAlO_3$ (100) single-crystal substrates, had a record high J_c for the Bi oxide system: $J_c\geq10^9$ A/m^2 at 77.3 K in high magnetic fields ($\geq$1T, $B\|a$-b plane),[19,20] and zero-field $J_c=10^{11}$ A/m^2 at 30 K.[21] In this paper we investigate the thermally activated electrical resistivity ρ in these high-quality Bi-2:2:2:3 thin films in magnetic fields applied parallel to the c axis. For a wide temperature and field range the resistivity shows clear Arrhenius behavior with an activation energy $U_0=A(1-T/T_c)/\sqrt{B}$, which is explained by the plastic motion of pinned flux liquid. Flux-creep theory predicts that B^*—defined by an electric field E criterion—is determined by U_0/kT, which leads to a temperature dependence of $\sqrt{B^*}=C(1/kT-1/kT_c)$. We have confirmed that this relation holds not only for B^* of our films but also for several reported B^* of Bi-2:2:2:3 samples measured by dc magnetization.

II. EXPERIMENT

Bi oxide thin films were deposited on $LaAlO_3$(100) single-crystal substrates by a MOCVD technique. The details of sample preparation were reported elsewhere.[19] The films used in this study, 60–80-nm-thick, were characterized by an x-ray diffractometer and found to be single-phase Bi-2:2:2:3 thin films with the c-axis oriented perpendicular to the film surface. The surface morphology examined by high-resolution scanning electron microscopy was very smooth without apparent grain boundaries and reflection high-energy electron diffraction (RHEED) pictures showed clear streak patterns, which indicates that these films are epitaxial films.

The electrical resistivity and E-J relation were measured by a four-probe dc method using 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 in an oxygen gas atmosphere [$p(O_2)=20$–30 Torr] at 300 °C to reduce the contact resistivity. Resistivity was measured as functions of magnetic field and temperature in a variable temperature insert, using a 14-T superconducting magnet. Specimen temperature was controlled by a AuFe-chromel thermocouple and a capacitance sensor with an accuracy of ±0.2 K.

III. RESULTS AND DISCUSSION

A. Thermally activated flux motion in Bi-2:2:2:3 films

It has been known that the resistive transitions of Bi-2:2:1:2 single crystals and thin films in magnetic fields

are well approximated by the thermally activated form $\rho(B,T)=\rho_0\exp(-U_0/kT)$.[22,23] Figure 1 shows the Arrhenius plots of the resistivity $\rho(B,T)$ of our Bi-2:2:2:3 film ($\sim$80 nm thick) measured as a function of temperature in fixed fields parallel to the c axis.[24] The critical temperature of the film was $T_c(R=0)\approx 86.5$ K with T_c(midpoint)≈ 100 K and T_c(onset)≈ 110 K. Except near $1/T_c$ where superconducting fluctuations set in, these plots are almost linear in a wide $1/T$ range. From the straight portion of the data ($\rho \leq 0.1\rho_n$, where ρ_n is the normal resistivity) the activation energy U_0 is calculated and plotted against B in the upper inset of Fig. 1, indicating that U_0 is proportional to $B^{-0.472}\approx 1/\sqrt{B}$. The $1/\sqrt{B}$ dependence of U_0 has been demonstrated in the analyses of $\rho(B,T)$ data in Bi-2:2:1:2 thin films,[23] although a $1/B$ dependence has widely been observed in YBCO.[15,25]

To confirm the $1/\sqrt{B}$ dependence of U_0, the resistivity ρ was measured as a function of B at various fixed temperatures of 30–80 K. Figure 2 shows Arrhenius-type plots of ρ vs $1/\sqrt{B}$ for the same sample as was used for Fig. 1, but that was a little degraded after several measurements, as $T_c(R=0)\approx 85.4$ K. A clear linear relationship between $\ln\rho$ and $1/\sqrt{B}$ for a wide T and B range confirms the $1/\sqrt{B}$ dependence of the activation energy:

$$\rho(B,T)=\rho_0\exp(-U_0/kT)=\rho_0\exp[-\alpha(T)/kT\sqrt{B}\,] .$$

The temperature-dependent factor α/k calculated from the straight lines shown in Fig. 2 is plotted as a function of temperature in the inset of Fig. 2. We can draw a straight line on the plot for $T\geq 45$ K. The intersection with the horizontal axis gives a value of $T=95$ K, which corresponds to the T_c of this film whose midpoint T_c is $\sim$100 K. At lower temperatures $T\leq 40$ K the α/k data points slightly deviate upward from the straight line. For our later analysis of the irreversibility field we approximate $\alpha(T)$ as $\sim(1-T/T_c)$. Then our experimental

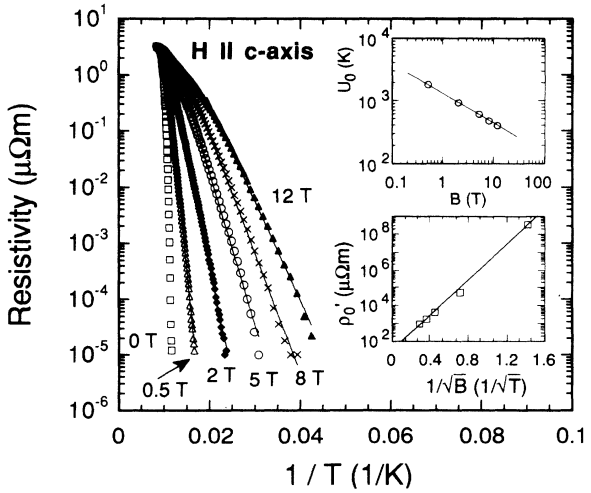

FIG. 1. Arrhenius plots of the resistivity $\rho(B,T)$ of an epitaxial Bi-2:2:2:3 film in the field parallel to the c axis. Upper inset: activation energy U_0 plotted against the applied field B. Lower inset: apparent prefactor ρ_0' plotted against $1/\sqrt{B}$ (see text).

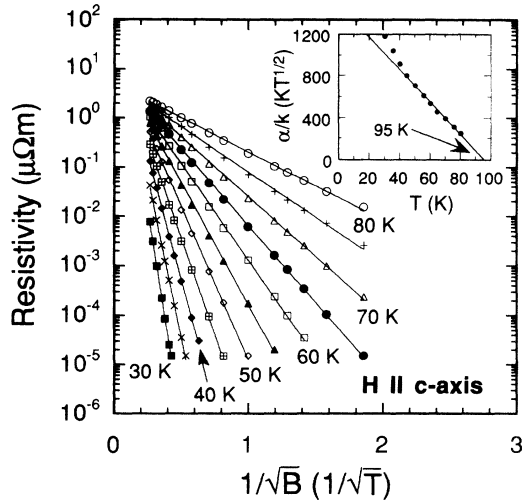

FIG. 2. Arrhenius-type plots of ρ vs $1/\sqrt{B}$ for Bi-2:2:2:3 film for temperatures $T=30$–80 K with an interval of 5 K, showing $\rho(B,T)=\rho_0\exp[-\alpha(T)/kT\sqrt{B}\,]$. Inset: calculated values of α/k plotted as a function of temperature.

$\rho(B,T)$ data are expressed as

$$\rho(B,T)=\rho_0\exp(-U_0/kT)$$
$$=\rho_0\exp[-A(1-T/T_c)/kT\sqrt{B}\,] . \quad (1)$$

The same B and T dependence of resistivity has been reported for Bi-2:2:1:2 epitaxial films, but the linear T dependence of the activation energy, $U_0\propto 1-T/T_c$, was observed only in a limited field range, $B\leq 1$ T.[23] Equation (1), which can be rewritten as

$$\rho(B,T)=[\rho_0\exp(A/kT_c\sqrt{B}\,)]\exp(-A/kT\sqrt{B}\,)$$
$$=\rho_0'\exp(-A/kT\sqrt{B}\,) ,$$

predicts that in a fixed magnetic field a linear relationship between $\ln\rho$ and $1/T$ holds. This was actually observed in Fig. 1, and the lower inset of Fig. 1 confirms the field dependence of the apparent prefactor $\rho_0'=\rho_0\exp(A/kT_c\sqrt{B}\,)$. That is, the activation energy plotted in the upper inset of Fig. 1 corresponds to the U_0 value at $T=0$ K. To our knowledge such a clear magnetic field and temperature dependence of the thermally activated dissipation as is shown in Figs. 1 and 2 has never been observed for Bi oxides.

Geshkenbein *et al.* proposed a model which gives a physical origin for the observed $1/\sqrt{B}$ dependence of U_0.[26] In their model U_0 is associated with the plastic deformation of the flux-line lattice at flux-line-lattice dislocations, analogous to the thermally activated motion of edge dislocations in crystals. U_0 is regarded as the energy required to create a double kink in the flux line:

$$U_0=2\varepsilon_1 a_0\approx(\Phi_0^2/2\pi\mu_0\lambda_{ab}\lambda_c)\ln\kappa\sqrt{\Phi_0/B} , \quad (2)$$

where ε_1 is the extra energy per unit length of the flux line along the CuO_2 plane, a_0 the flux-line-lattice spacing, Φ_0 the flux quantum, λ_{ab} and λ_c the penetration depth when the screening currents are flowing in the a-b plane

and along the c axis, and κ the Ginzburg-Landau (GL) parameter.[26,27] This model also explains why the temperature dependence is proportional to $1-T/T_c$ near T_c, since $\lambda \propto (1-T/T_c)^{-1/2}$. Later Vinokur *et al.*[28] have extended this model to the plastic motion of pinned flux liquid, incorporating the idea of vortex glass-liquid phase transition[5-7].

Recently Li *et al.*[29] derived the values of $\kappa \approx 170$ and $\lambda_{ab}(0)=194$ nm for c-axis-oriented Bi-2:2:2:3 samples in their analyses of the reversible magnetization data with the Hao-Clem theory.[30] Their value of $\lambda_{ab}(0)$ is close to the previous value of $\lambda_{ab}(0)=232$ nm reported by Shilling, Hulliger, and Ott.[31] However, there have been no reliable estimate of $\lambda_c(0)$ or $\gamma=(m_c/m_{ab})^{1/2}=\lambda_c/\lambda_{ab}$ for the Bi-2:2:2:3 phase, where m_c and m_{ab} are the Ginzburg-Landau superconducting effective masses for pair motion along the c direction and in the a-b plane. For Bi-2:2:1:2 single crystals Farrel *et al.* reported the value of $\gamma=55$ from magnetic torque measurement,[32] and recently Kleiner *et al.* reported $\gamma \approx 50$ for O_2 annealed samples and $\gamma \leq 280$ for Ar annealed samples.[33] If we use the values $\kappa=170$, $\lambda_{ab}(0)=200$ nm, and $\gamma=50$ for our Bi-2:2:2:3 film, the activation energy of Eq. (2) is calculated as $U_0(1T)=4580$ K. This value is more than three times larger than the observed value of $U_0(1T)=1320$ K (Fig. 1).

The calculation of flux-line energy in Eq. (2) is based on the London theory for an anisotropic large κ superconductor with homogeneous structure.[27] However, the above parameters used for the estimate of U_0 result in unphysically short GL coherence length $\xi_c(0) \approx 0.02$ nm. This suggests that we need to take into account the layer structure of Bi oxides explicitly. According to Tachiki and Takahashi[34] Bi oxides are comprised of the superconducting CuO_2 layers and other blocking layers which are normal or weakly superconducting with lower order parameter. The flux lines along the CuO_2 planes are considered to enter into these blocking layers because the flux-line energy is lower, and the stepwise flux penetration with many kinks is expected.[34] This is confirmed by the angular dependence of $J_c(B,\theta)$ determined only by the magnetic field component along the c direction, $J_c(B,\theta)=J_{c1}(B\sin\theta)$.[21,35] In case of the thermally activated plastic motion the extra flux lines along the CuO_2 planes are considered to enter into these blocking layers, and their flux-line energy should be smaller than the estimate based on Eq. (2). Koyama *et al.* calculated the lower critical field $B_{c1}^{\parallel}$ (the flux-line energy divided by Φ_0) along the CuO_2 planes using a new Ginzburg-Landau model in which the spatial-dependent coefficients in the GL free energy are assumed to describe the layer structure.[36] Their numerical calculation shows that $B_{c1}^{\parallel}$ is lower than calculated by the continuous London model[27] and the temperature dependence of $B_{c1}^{\parallel}$ has an upturn curvature as temperature decreases [Eq. (45), and Figs. 6 and 7 in Ref. 36], similar to the temperature dependence of α/k in the inset of Fig. 2. This upturn curvature is interpreted to be a characteristic feature in proximity-induced superconducting systems.[36]

Although it is incorrect to use the continuous London model for the calculation of ε_1 of extremely anisotropic Bi oxides [Eq. (2)], Geshkenbein *et al.*'s double-kink model explains well the observed $1/\sqrt{B}$ dependence of the activation energy, $U_0=2\varepsilon_1 a_0$. We note that their model is not valid if superconducting planes are completely decoupled and the material is transparent for a magnetic field parallel to the CuO_2 planes.[37] We do not think that superconducting planes are completely decoupled, since very recently we have observed in our Bi-2:2:2:3 thin films the finite-temperature vortex-glass transition,[38] which is not expected in the strictly two-dimensional (2D) system.[5,39] Recent magnetization measurement on heavy-ion-irradiated Bi-2:2:1:2 crystals presents experimental evidence for the line features of the vortices against the interpretation of vortex properties in terms of decoupled 2D pancakes.[40] We therefore attribute the thermally activated dissipation in our Bi-2:2:2:3 films [Eq. (1)] to Geshkenbein *et al.*'s double-kink model. In addition, from many experimental results described later the region where finite resistivity is observed is supposed to be in the vortex-liquid regime. Our results are attributed to the plastic motion of pinned flux liquid.[28]

B. Irreversibility fields of Bi oxides

We investigated the irreversibility field B^* by measuring E-J relation as functions of magnetic field and temperature. Figure 3 shows the E-J relation in a region near B^*. We can see the transition from the zero-resistivity region where there is a finite critical current density J_c to the resistive region where J_c vanishes. The transition is sharp enough to define the irreversibility field: for our Bi-2:2:2:3 thin films having zero field $J_c \geq 10^{10}$ A/m^2 at 30–40 K,[21] we define B^* as the field at which the induced electric field becomes $E=1$ μV/cm with a current density of 10^7 A/m^2, because the transition from the linear to nonlinear E-J relation seems to occur near this point.[18,20] We note that $\rho(B,T)$ data shown in Figs. 1 and 2 are taken above B^* where there is a finite observable zero-bias resistance, $(dE/dJ)_{J=0}>0$. Figure 4 shows the T dependence of B^* values for our Bi-2:2:2:3 films and other B^* values reported so far for Bi oxide superconductors. The specimens are magnetically

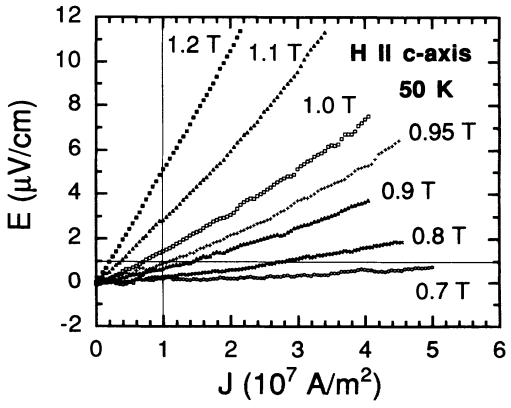

FIG. 3. E-J relation measured in a region near B^*.

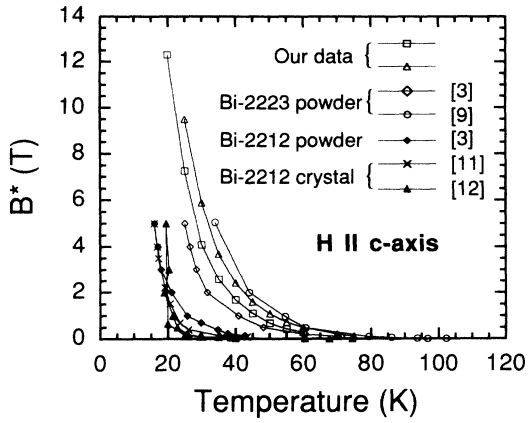

FIG. 4. Temperature dependence of resistively measured B^* of our Bi-2:2:2:3 films and the reported B^* values. Open symbols denote Bi-2:2:2:3 samples, and solid symbols and crosses ($\times$) denote Bi-2:2:1:2 samples.

oriented Bi-2:2:1:2 and Bi-2:2:2:3 powders encapsulated in epoxy[3,9,10] and Bi-2:2:1:2 single crystals;[11,12] the B^* values were measured by dc magnetization. Since these measurements are based on the intragrain current density, an ambiguity originating from weak links at grain boundaries is excluded.

We now discuss the temperature dependence of B^* using the flux-creep model.[41–43] In this study we assume a sinusoidal spatial variation of pinning potential,

$$U(X)=\tfrac{1}{2}U_0\cos(\pi X/X_p)-JBV_aX\ ,$$

where V_a is the activation volume, and X_p is the effective length of the potential well.[42,44] The electric field induced by the thermally assisted flux motion is given by[18,43,44]

$$E=2E_0\exp\left[-U_0\left[(1-j)^{3/2}+\frac{\pi}{2}j\right]\Big/kT\right]\times\sinh(\pi U_0 j/2kT)\ , \tag{3}$$

where $j=J/J_0$, and J_0 is the critical-current density when there is no thermal activation. In the region near the irreversibility field where $j\ll 1$, Eq. (3) reduces to

$$E=2E_0\exp(-U_0/kT)\sinh(\pi U_0 J/2kTJ_0)\ , \tag{4}$$

because $(1-j)^{3/2}+(\pi/2)j\approx 1$. We note that in the original Kim-Anderson theory—which assumes a linear J dependence of the effective activation barrier $U_{\rm eff}=U_0(1-j)$ when $U_0 j/kT$ is not small—a similar form

$$E=2E_0\exp(-U_0/kT)\sinh(U_0J/kTJ_0)$$

is obtained.[41,43] In the thermally assisted flux-flow (TAFF) region[45] where $U_0J/kTJ_0\ll 1$, Eq. (4) reduces to $E=(\pi E_0U_0/kTJ_0)J\exp(-U_0/kT)$, since $\sinh(x)\approx x$ for small x. We then obtain

$$\rho=E/J=(\pi E_0U_0/kTJ_0)\exp(-U_0/kT)\ . \tag{5}$$

This equation corresponds to the empirical relationship, Eq. (1). Equations (4) and (5) show that in the region near B^* the electric field E is dominated by the term $\exp(-U_0/kT)$ when $U_0\gg kT$, which is satisfied in our Bi-2:2:2:3 films except near T_c (Figs. 1 and 2). Therefore, B^*—defined by an electric-field criterion—is determined by U_0/kT. At any given temperature, B^* is reached when U_0/kT takes a specific value corresponding to the electric-field criterion E_c: $B=B^*$ when $U_0/kT=\ln(K/E_c)$, where K is a constant. Substituting U_0 by $A(1-T/T_c)/\sqrt{B^*}$ [Eq. (1)], we obtain the relation

$$\sqrt{B^*}=C(1/kT-1/kT_c)\ . \tag{6}$$

We see in Fig. 5 that for our Bi-2:2:2:3 film and for Bi-2:2:2:3 oriented powders there is a clear linear relationship between $\sqrt{B^*}$ and $1/T$, and the intersections of the straight lines with the horizontal axis give values of $T=94$–112 K, corresponding to the T_c's of these samples.[46] It is seen that Eq. (6) expresses the temperature dependence of the irreversibility field of the Bi-2:2:2:3 superconductors very well. Equation (6) is the consequence of the thermally activated dissipation expressed by Eq. (1), which is attributed to the thermally activated plastic motion of pinned flux liquid.[28] This result therefore suggests that the plastic motion of pinned flux liquid is generally observed in Bi-2:2:2:3 specimens.

For the Bi-2:2:1:2 samples, however, the situation is uncertain. Kucera *et al.* reported thermally activated behavior similar to Eq. (1) for Bi-2:2:1:2 epitaxial films: as for the low-resistance regime near B^*, the linear T dependence of $U_0\propto 1-T/T_c$ was only observed in the low-field range ($B\le 1$ T), and at higher fields a nonlinear behavior, attributed to a possible 2D to three-dimensional (3D) crossover, is observed.[23] If only low-field ($B^*\le 1$ T) data are used for curve fitting, we can draw a straight line and obtain a somewhat reasonable value of $T_c\approx 70$ K for the data of Ref. 3 for Bi-2:2:1:2 oriented powders (Fig. 5). But the data of Refs. 11 and

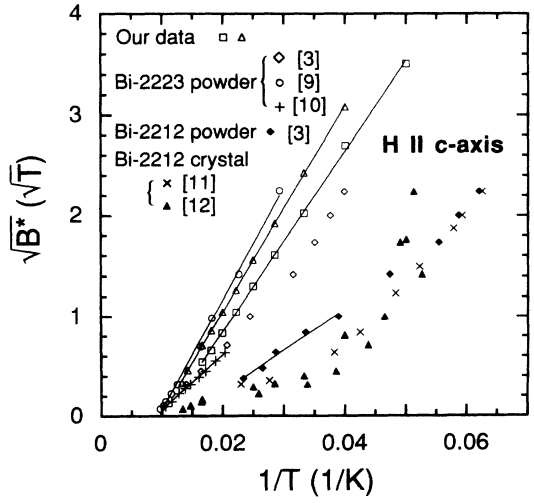

FIG. 5. $\sqrt{B^*}$ vs $1/T$ plots for our Bi-2:2:2:3 thin films and for reported B^* data. Open symbols and pluses (+) denote Bi-2:2:2:3 samples, and solid symbols and crosses ($\times$) denote Bi-2:2:1:2 samples.

12 for Bi-2:2:1:2 single crystals cannot be fitted to Eq. (6). In fact, the field and temperature dependences of U_0 in Bi-2:2:1:2 single crystals are unclear: Palstra *et al.* reported a temperature-independent activation energy $U_0 \sim B^{-\alpha}$ with $\alpha \approx \frac{1}{6}$ ($B < 3$ T) and $\alpha \approx \frac{1}{3}$ ($B > 3$ T),[22] but Krause, Shi, and Datars reported that $U_0 \sim B^{-1/3}$ for $B \leq 1.6$ T assuming the T dependence of $\sim[1-(T/T_c)^2]^{3/2}$.[47] If we want to observe a temperature dependence of B^* similar to Eq. (6) for Bi-2:2:1:2, we need to study both the thermally activated dissipation and B^* for the same sample.

Since the thermally activated dissipation depends on the pinning strength and the anisotropy of the system,[23] the different behavior of Bi-2:2:1:2 and Bi-2:2:2:3 is attributed to the difference of those properties. We have stated that the thermally activated plastic motion of pinned flux liquid,[28] to which our experimental results are attributed, cannot be applied to 2D pancake vortices with completely decoupled superconducting planes.[37] The fact that the plastic motion of pinned flux liquid is generally observed in Bi-2:2:2:3 suggests that Bi-2:2:2:3 specimens have enough pinning and that the simple 2D picture is not the case for wide temperature and magnetic-field range.

There have been several reports that suggest a possible magnetic phase transition well below the irreversibility line in Bi-2:2:1:2 samples.[48,49] Recently Safar *et al.* observed a clear crossover from high-temperature Arrhenius behavior to a critical region associated with the low-temperature 3D vortex-glass phase transition in Bi-2:2:1:2 single crystal.[50] Wang *et al.* observed the vortex-glass transition in Tl-2:2:2:3 thin films, showing that the phase boundary is not the irreversibility line.[51] Very recently we have also observed the vortex-glass–liquid transition in our Bi-2:2:2:3 thin films and confirmed that the vortex-glass transition line is well below the irreversibility line as determined in this paper.[38] Taking these observations into consideration, the thermally activated dissipation shown in Figs. 1 and 2 and the T dependence of B^* [Eq. (6)] are the phenomena observed *above* the vortex-glass–liquid transition line. We believe, therefore, that they correspond to the plastic motion of pinned flux liquid.[28]

IV. SUMMARY

Thermally activated resistivity in epitaxial $Bi_2Sr_2Ca_2Cu_3O_x$ thin films can be well approximated by an activation energy $U_0 = A(1-T/T_c)/\sqrt{B}$. We have observed a clear temperature dependence of the irreversibility field, $\sqrt{B^*} = C(1/kT - 1/kT_c)$, as predicted from flux-creep theory when B^* is defined by an electric-field criterion. It is suggested that the irreversibility line originates from the thermally activated plastic motion of pinned flux liquid.

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