Dependence of activation energy upon magnetic field and temperature in $YBa_2Cu_3O_{7-\delta}$ epitaxial thin film

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The broadening of the resistive transition in YBa₂Cu₃O_{7- δ} epitaxial thin film was measured in various applied magnetic fields. The irreversibility line and its angular dependence were obtained. The temperature and magnetic field dependence of the activation energy $U_0(H,T) \propto U_0[1-T/T_c(0)](H_0/\varepsilon_{\theta}H)^{2/3}$ were obtained from the experimental results, which is consistent with the expression of the activation energy for thermally excited flux creep deduced from the irreversibility line and Arrhenius law except for low field data in which the prefactor of the Arrhenius law has little deviation from that in high fields. [S0163-1829(99)10301-1]

I. INTRODUCTION

In high- T_c oxide superconductors, the field-temperature (H-T) phase diagram is complicated due to the high transition temperature, short coherence length, layered structure, and high anisotropy. The phase transition at the upper critical field H_{c2} is smeared out and it is argued that a true phase transition occurs at the irreversibility line (IL). The IL was believed to be a vortex lattice melting line or a vortex glass transition, depending on the strength of the disorder in superconductors. For example, in high quality untwinned crystals of YBa₂Cu₃O_{7- δ} (YBCO), the phase transition at the IL determined by a sharp "kink" is a first-order melting transition from the Abrikosov vortex lattice state to a vortex liquid state,¹⁻⁷ whereas in thin films, it exhibits a secondorder transition from a vortex glass state to a vortex liquid state⁸⁻¹³ and the IL is generally determined by the criterion of zero resistance in the measurements of the resistive transition. The first-order phase transition from the vortex solid to the vortex liquid in the untwinned YBCO crystal can be suppressed by the introduction of point defects.¹⁴ Using twodimensional electron-gas Hall-sensor arrays, Majer et al.¹⁵ measured the local magnetization loops in a Bi₂Sr₂CaCu₂O₈ single crystal and showed that the IL in Bi₂Sr₂CaCu₂O₈ is due to geometrical barriers at high temperatures and surface barriers at intermediate temperatures. Their results clearly demonstrated that the IL and melting stem from different and unrelated physical mechanisms. The thermally activated flux motion behavior is commonly observed just above the IL in disorder superconductors. The resistivity broadening was

thought to be because of the thermally activated flux motion¹⁶⁻²² and can be fitted by Arrhenius law for the resistivity below $0.01\rho_n$,^{23–25} where ρ_n is the normal state resistivity. A common intersection in the Arrhenius plot showed that the temperature dependence of resistivity in various magnetic fields could be described by a single prefactor ρ_0 .^{16,26} The activation energy of magnetic flux motion U_0 which is a function of the temperature and magnetic field, can be evaluated from Arrhenius plots. In this paper, the broadening of the resistive transition in YBCO thin film was measured in various magnetic fields and at different angles between the direction of the magnetic field and *ab* plane. The results show the angular dependence of the IL coincides with the effective mass model. The activation energy as a function of the magnetic field and temperature is obtained from both the experimental results and the theory of thermally excited flux creep.

II. EXPERIMENTAL

A high quality *c*-axis-orientated YBCO epitaxial thin film was prepared by high-pressure (p=3 mbar) dc sputtering in a pure oxygen atmosphere from a planar and stoichiometric YBCO target ($\Phi=35$ mm) onto a heated SrTiO₃ (100) substrate as reported elsewhere.²⁷ The x-ray diffraction showed that no impurity phase peaks were present and the full width at half maximum (FWHM) of the rocking curve for the (005) diffraction peak was $\leq 0.3^{\circ}$, indicating a very good epitaxial *c*-axis orientation. The ac-susceptibility measurement as a function of temperature exhibited a superconducting transi-

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tion temperature of T_c =91.6 K, and the width of the resistive transition ΔT_c was <0.3 K which was defined as a temperature interval from 90% χ'_{ac} to 10% χ'_{ac} . This was consistent with the electric measurement. The critical current density J_c was 3.8×10^6 A/cm² at 77.3 K in zero magnetic field.

The film was patterned into a narrow bridge 20 μ m in width, 100 μ m in length, and 300 nm in thickness. The silver leads were attached by indium solder on Ag terminals deposited on the film. The electric measurements were performed using a standard four-probe technique. The film was held on a sample holder consisting of a worm-gear driving system. Using this system, the relative angle between the direction of the external magnetic field and *ab* plane of the film can be conveniently adjusted to a resolution of 0.1°. The magnetic field up to 10 T was supplied by a water-cooled magnetic system. The temperature of the sample was measured by calibrated Rh-Fe resistance thermometer and the magnetic field effect on the temperature was corrected.

III. EXPERIMENTAL RESULTS

The temperature dependence of the resistivity of YBCO epitaxial thin film was measured for the configurations of (a) $H \perp ab$ plane and $H \perp i$, (b) $H \parallel ab$ plane and $H \perp i$, (c) $H \parallel ab$ plane and $H \parallel i$ in magnetic field up to 8 T, and (d) H = 4 T with various angle θ and $H \perp i$, where i is the in-plane current density. Figure 1(a) shows Arrhenius plots of the inverse temperature dependence of the reduced resistivity for the $H \perp j$, $H \perp ab$ plane and $H \parallel ab$ plane, respectively. The experimental results show that the $\rho(T)$ transition is very steep at zero magnetic field, and there exists a broadening for any configuration under application of magnetic field. At the same magnetic field strength, the width of the broadening for the $H \perp ab$ plane is much larger than that for the $H \parallel ab$ plane. However, there is only a slight difference of $\rho(T)$ broadening between $H \parallel j$ and $H \perp j$ for the $H \parallel ab$ plane. This means that the broadening of the resistive transition depends on the relative direction between magnetic field and crystal axis, rather than on the relative direction between magnetic field and current. Figure 1(b) shows Arrhenius plots of $\rho(T)$ broadening for various angles θ at H=4 T and $H\perp j$. With the increasing of the angle θ the broadening of the resistive transition increases rapidly at low angles and slowly at high angles. The relation of the broadening and the angle θ is determined by the angular dependence of the irreversibility field [Eq. (1) in Sec. III A].

A. Irreversibility line

Figure 2 shows the dependence of $\varepsilon_{\theta}H$ upon $T_c(H,\theta)$ under various magnetic fields and various angles with the criterion of zero resistance (the voltage resolution is 10 nV found by a nanovoltmeter in our measurements). The critical field defined by the zero resistance criterion was generally considered to be the irreversibility field,²⁸ i.e., the *H*-*T* relation shown in Fig. 2 may be believed to be the irreversibility line. This relation can be approximately expressed by an equation [the relation of $\ln(\varepsilon_{\theta}H)$ versus $\ln(1-t)$ shown in the inset of Fig. 2]:



FIG. 1. Arrhenius plots of YBCO thin film. The inset of (b) shows the angle θ .

$$\varepsilon_{\theta} H = H_0 (1 - t)^{\mu}. \tag{1}$$

Here, $\varepsilon_{\theta} = (\sin^2\theta + \varepsilon^2 \cos^2\theta)^{1/2}$ is the anisotropy-angledependence parameter²⁹ and $\varepsilon = \frac{1}{6}$. $t = T_c(H, \theta)/T_c(0)$ is the reduced temperature and $T_c(0) = 91.6$ K. $H_0 = 87$ T is the irreversibility field in the *c*-axis direction at zero temperature. μ is equal to about 1.5. In fact, $\varepsilon_{\theta}H$ is an effective field. In addition, $\varepsilon_{\theta}H = H$ for the $H \perp ab$ plane, $\varepsilon_{\theta}H = \varepsilon H$ for the $H \parallel ab$ plane. This result indicates that the angular dependence of the irreversibility line is consistent with the effective mass model.

B. $U_0(H,T)$

Figure 3 shows $U_0(T,H)/k_B = -T \ln(\rho/\rho_0)$ as a function of temperature (*T*) for the magnetic field configuration of the $H \parallel c$ axis, where k_B is the Boltzmann constant. This figure shows that when the temperature is in the regime of the flux creep, the activation energy decreases linearly with increasing temperature at a fixed applied magnetic field.

The relation between $[U_0(T,H)/k_B]/[1-T/T_c(0)]$ = $-T \ln(\rho/\rho_0)/[1-T/T_c(0)]$ and T is shown in Fig. 4, where



FIG. 2. The dependence of $\varepsilon_{\theta}H$ upon $T_c(H,\theta)$ with the criterion of $\rho=0$. The inset shows the relation of $\ln(\varepsilon_{\theta}H)$ and $\ln[1-T_c(H,\theta)/T_c(0)]$. The solid line shows the best fit.

 $T_c(0) \cong 91.6$ K is the temperature where $U_0(T,H) = 0$ by the linear extrapolating of $U_0(T,H)$. Figure 4 indicates $U_0(T,H)/k_B \propto 1 - T/T_c(0)$ when temperature is in the regime of the flux creep. Moreover, the relationship between $U_0(H)$ and H can also be obtained as shown in Fig. 5. With the increasing of the magnetic field, $U_0(H)$ decreases rapidly in a small field and slowly in a high field. In this way, one can obtain $U_0(H)$ in any magnetic field configuration.

There is a similar magnetic field dependence of $U_0(H)$ for three typical configurations of magnetic field. However, the $U_0(H)$ for the $H \perp ab$ plane is smaller than that for the $H \parallel ab$ plane at the same strength of magnetic field. The inset of Fig. 5 plots the relation of $U_0(H)$ versus $(H_0/\varepsilon_{\theta}H)^{1/\mu}$ and can be described by the following expression:





FIG. 3. The dependence of $U_0(T,H)/k_B = -T \ln(\rho/\rho_0)$ upon temperature.



FIG. 4. The temperature dependence of $[U_0(T,H)/k_B]/[1-T/T_c(0)]$.

where μ and H_0 are the parameters obtained from the irreversibility line. Equation (2) gives the angular dependence of activation energy on the magnetic field and is in agreement with the description of the effective mass model.

IV. DISCUSSION

Palstra *et al.*^{16,17} reported the broadening of the resistive transition in magnetic field and indicated that there is a common intersection of the extended lines of $\ln(\rho)$ vs 1/T in various magnetic fields for single crystal Bi₂Sr₂CaCu₂O_y. Cao *et al.*²⁶ obtained similar results in their study on the dissipation in weak links of the Tl-Ba-Ca-Cu-O superconductor. In this work, we observed that the common intersection is in agreement with the point $[T_c^{-1}(0), \ln \rho_0]$ for YBCO thin film and $T_c(0)=91.6$ K, $\rho_0=(401\pm5)\rho_n$; the deviation is mainly from low field data. It is speculated that the effect and range of pinning may change dramatically due to



FIG. 5. The dependence of $U_0(H)$ upon $\varepsilon_{\theta}H$. The inset plots the relation of $U_0(H)$ and $(H_0/\varepsilon_{\theta}H)^{2/3}$. The solid line is the best fit.

the rapid change in the superconducting coherence length as the temperature of the sample approaches $T_c(0)$,⁷ and this may result in the deviation of ρ_0 . Furthermore, superconducting fluctuations which may change the ρ_0 will also be important at low fields and near $T_c(0)$. Qiu *et al.*³⁰ and Sun *et al.*³¹ observed a similar result in YBCO thin films and (Bi, Pb)₂Sr₂CaCu₂O_x silver-sheathed tape, respectively. These results imply that the temperature dependence of resistivity in various fields can be described by a single prefactor ρ_0 .

Generally, the IL can be written as the function

$$H = H_0 [1 - T_{\rm irr} / T_c(0)]^{\mu}, \qquad (3)$$

where T_{irr} is the irreversibility temperature. However, the so-called $\rho = 0$ as the criterion of the IL is restricted by the sensitivity of the instruments. We suppose that the lowest value of ρ is equal to e^{-n} . According to the above discussion, we know that points $(T_{irr}^{-1}, -n)$ and $[T_c^{-1}(0), \ln \rho_0]$ are on the Arrhenius line and its extension. Therefore, the Arrhenius line can be described by

$$(\ln \rho_0 - \ln \rho) / [T_c^{-1}(0) - T^{-1}]$$

= $(\ln \rho - \ln e^{-n}) / (T^{-1} - T_{irr}^{-1}).$ (4)

Let $\ln \rho_0 = m$, the solution of Eq. (4), have the form

$$\rho = \rho_0 \exp\{-(m+n)(T_{\rm irr}/T)[T-T_c(0)]/[T_{\rm irr}-T_c(0)]\}.$$
(5)

Using the relation between T_{irr} and H of Eq. (3), the resistivity expression of Eq. (5) becomes

$$\rho = \rho_0 \exp\{-(m+n)T_c(0)[1-T/T_c(0)] \\ \times [(H_0/H)^{1/\mu} - 1]/T\}.$$
(6)

Comparing Eq. (6) with the Arrhenius law $\rho = \rho_0 \exp \left[-U_0(H,T)/T\right]$, it is obtained that the dependence of activation energy upon the magnetic field and temperature is

$$U_0(H,T) = U_0(0)(1-t)[(H_0/H)^{1/\mu} - 1], \qquad (7)$$

where $t = T/T_c(0)$, $U_0(0) = (m+n)k_BT_c(0)$. The order of (m+n) is 10 and that of $T_c(0)$ is 100, so the order of $U_0(0)$ is 1000 k_B .

Taking Eq. (6) into the logarithm then differentiating it with respect to T^{-1} , we can obtain

$$\partial \ln(\rho) / \partial T^{-1} = -(m+n)k_B T_c(0) [(H_0/H)^{1/\mu} - 1].$$
 (8)

Equation (8) indicates that the slope of the Arrhenius line is irrelevant to the temperature and agrees with the experimental results. Comparing Eq. (7) with Eq. (8), we have

$$U_0(H,T) = -(1-t)\partial \ln(\rho) / \partial T^{-1}.$$
 (9)

Equation (9) shows the dependence of the slope of the Arrhenius line upon activation energy $U_0(H,T)$. Generally, because $H_0/H \ge 1$ for high- T_c superconductors, $U_0(H,T)$ can be approximately expressed as

$$U_0(H,T) = U_0(0)(1-t)(H_0/H)^{1/\mu},$$
(10)

i.e., $U_0 \propto H^{-1/\mu} \approx H^{-2/3}$. Equation (10) is in agreement with the experimental results. The similar expression of $U_0 \propto (1-t)H^{-0.7\pm0.1}$ corresponds to an electron irradiated untwinned YBCO crystal reported by Fendrich *et al.*¹⁴

Geshkenbein³² and Vinokur³³ have made a theoretical attempt at predicting the $U_0 \propto (1-t)B^{-1/2}$ behavior, based on the model of plastic flux creep ascribing the dissipation to the plastic shear of dislocations in weakly pinning vortex liquid. The relation between the activation energy and the temperature coincided with our results. But, the magnetic field dependence of $U_0 \propto H^{-2/3}$ that we obtained is different from that of their theory, i.e., $U_0 \propto B^{-1/2}$.

Kadowaki et al.²⁵ proposed the expression

$$U_0(H,T) = U_0(H)(1-t^2)^{3/2}$$
(11)

and Kim et al.21 proposed the expression

$$U_0(H,T) = U_0(H)(1-t)^q.$$
 (12)

Inserting Eqs. (11) and (12) into the Arrhenius law and making a simple algebra operation, we have an expression corresponding to Eqs. (11) and (12), respectively,

$$\partial \ln(\rho) / \partial T^{-1} = -U_0(H)(1-t^2)^{3/2}(1+2t^2)/(1-t^2),$$
(13)

$$\partial \ln(\rho) / \partial T^{-1} = -U_0(H)(1-t)^q [1+(q-1)t] / (1-t).$$
(14)

Equations (13) and (14) show that $\partial \ln(\rho)/\partial T^{-1}$ is a function of temperature except for q=1 in Eq. (14). This contradicts the linear relationship of $\ln \rho$ and 1/T when $\rho < 0.01\rho_n$. This implies that, although the linear relationship of $\ln \rho$ and 1/T is only an approximation for YBCO when $\rho < 0.01\rho_n$, the activation energy may not be deduced from the condensation energy, i.e., a simple equation of $U_0 = (1/2) \pi H^2(T) a^{\alpha} \xi^{\beta}$, where *a* is the parameter of flux lattice and $\alpha + \beta = 3$.

In summary, we analyzed the broadening of the resistive transition of a YBCO thin film by means of thermally excited flux creep and found that the activation energy U_0 depends on $H^{-2/3}$ and increases linearly with decreasing temperature. The temperature and magnetic field dependence of the activation energy has also been deduced based on the irreversibility line and the Arrhenius law under an approximation of a single prefactor in the Arrhenius law. The angular dependence of both the activation energy and the irreversibility line was obtained based on experimental results. The anisotropy of both the activation energy and the irreversibility field coincides with the scaling of the upper critical field by the effective mass model.

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