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Angular dependences of the thermodynamic and electromagnetic properties of the high- T_c superconductors in the mixed state

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The dependence upon the applied field **H** of the Gibbs free-energy difference ΔG between the mixed and normal states of a high- κ anisotropic type-II superconductor is examined; ΔG is found to be a function only of the reduced field $h = H/H_{c2}(\theta, \phi)$ for $H \gg H_{c1}$. Other thermodynamic quantities such as the magnetization and specific heat show similar behaviors. The same scaling property can also be observed for the magnetoresistivity, critical current density, and flux-line-lattice melting temperature of the high- T_c superconductors.

The high- T_c superconductors (HTSC) are characterized by not only their high transition temperatures, but also by (a) large anisotropy due to their layered structures and (b) extreme type-II behavior [Ginzburg-Landau (GL) parameter $\kappa \gg 1$].¹⁻³ Because of the anisotropy, the response of a sample to an external magnetic field H depends generally not only on the magnitude but also on the orientation of H. For example, we expect that the response of a sample in a field of 1 T parallel to the c axis is different from that in a field of the same magnitude but parallel to the *ab* plane; this difference is closely related to the angular dependence of the upper critical field H_{c2} . As will be shown in this paper by using the anisotropic GL theory, 4^{-7} the dependence upon H of the Gibbs free-energy-density difference ΔG between the mixed and normal states of a high- κ anisotropic type-II superconductor is, to a good approximation, a function only of the reduced field $h = H/H_{c2}(\hat{\mathbf{H}})$ for $H \gg H_{c1}$ $(H_{c1}$ is the lower critical field), so that the angular dependence is contained in H_{c2} . This property, which we may call the scaling property (in the sense that H_{c2} plays the role of measuring scale for H), also exists for other thermodynamic quantities such as the magnetization M, entropy difference ΔS , and specific heat difference ΔC (except that M, being a vector, has additional angular dependence), which are derivable from ΔG . Various experimental data suggest that this scaling property also holds in the fluctuation region for HTSC.^{8,9} Interestingly, the same scaling property can also be observed in the magnetoresistivity data of HTSC for low transport current densities;^{10–19} it suggests that this scaling property also may be observed for other quantities, and we find that the critical current density j_c (Refs. 20 and 21) and flux-line-lattice melting temperature T_m (Ref. 22) show the same scaling behavior.

A usual and simple approach to describe the thermodynamic properties of HTSC is to make use of the GL theory,¹⁻³ generalized to the anisotropic case by introducing an effective mass tensor m_{ij} .⁴⁻⁷ One characteristic of the GL theory is that it can be formulated in dimensionless form,¹⁻³ for which the energy is measured in units of $H_c^2/4\pi$ and magnetic field in units of $\sqrt{2}H_c$, so that the temperature T dependence of a superconductor is contained in the normalization factors such as $H_c^2/4\pi$ and $\sqrt{2}H_c$. Therefore, the Helmholtz free-energy density for the mixed state of an anisotropic superconductor containing the average magnetic flux density **B** can be written generally as

$$F = F_{s0}(T) + [H_c^2(T)/4\pi](\frac{1}{2} + B'^2) -[H_c^2(T)/8\pi]f(\mathbf{B}';\kappa;m_1:m_2:m_3),$$
(1)

where $F_{s0}(T)$ is the free-energy density of the Meissner state ($\mathbf{B} = 0$), $\mathbf{B}' = (B', \theta, \phi)$, $B' = B/\sqrt{2}H_c$ (θ and ϕ are the polar and azimuthal angles of \mathbf{B}), κ is the mean GL parameter, m_i (i = 1, 2, 3) are the principal values of m_{ij} ; f is a dimensionless function of \mathbf{B}' involving the parameter κ and the ratios $m_1 : m_2 : m_3$, and varies from 1 to 0 as B varies from 0 to B_{c2} . For example, Abrikosov's result^{2,3} for high fields ($H_{c2} - H \ll H_{c2}$) and the anisotropic version of Ref. 5, as well as the results of Refs. 6, 7, and 23, which are valid for lower fields, satisfy Eq. (1). We find from previous results^{2,3,5-7,23} the useful property that

$$f(\mathbf{B}';\kappa;m_1:m_2:m_3) \simeq f(b) \tag{2}$$

when $\kappa \gg 1$ and $H \gg H_{c1}$, where $b = B'/\tilde{\kappa} = B/B_{c2}(\theta,\phi)$, $B_{c2} = \sqrt{2}H_c\tilde{\kappa}$, $\tilde{\kappa} = \kappa/\sqrt{m(\theta,\phi)}$, and $m(\theta,\phi) = (m_1\cos^2\phi + m_2\sin^2\phi)\sin^2\theta + m_3\cos^2\theta$.^{4,5} For example, when $\kappa \gg 1$, the Abrikosov high-field result for F (Refs. 2, 3, and 5) can be expressed in the form of Eq. (1), where f can be approximated as

$$f = (1-b)^2 / \beta_A, \tag{3}$$

where $\beta_A = 1.16$ (Refs. 2 and 3) is independent of the orientation of \mathbf{H} ;²⁴ for intermediate fields ($H_{c1} \ll H \ll H_{c2}$) the result of Refs. 6, 7, and 23 can be approximated by

$$f = 1 - \eta_1 b \ln(\eta_2/b), \tag{4}$$

where η_1 and η_2 are only weakly field (*h*) dependent and can be roughly approximated by $\eta_1 \simeq 0.77$ and $\eta_2 \simeq 1.44/e = 0.53$.²³

We want to derive the mixed-state Gibbs free-energy density G, which has T and \mathbf{H} as independent variables.

5854

Using $G = F - \mathbf{H} \cdot \mathbf{B}/4\pi$ and $\mathbf{H} = 4\pi (\partial F/\partial \mathbf{B})_T$ we find

$$\Delta G = -[H_c^2(T)/8\pi]g(h) \tag{5}$$

for $\kappa \gg 1$ and $H \gg H_{c1}$, where $\Delta G = G - G_{nH}$, $G_{nH} = F_{s0}(T) + (H_c^2 - H^2)/8\pi$ is the normal-state Gibbs freeenergy density in **H**, and

$$g = f - 2(-4\pi M')^2 \simeq f(h);$$
 (6)

we have used the fact that $(-4\pi M')^2/f \sim \kappa^{-2} \ll 1$, and that H and B are much greater than $-4\pi M$, so that $\mathbf{H} \simeq \mathbf{B}$ and $h \simeq b$. Note that the conditions $\kappa \gg 1$ and $H \gg H_{c1}$, which are equivalent to $\kappa^2 b \gg 1$, can be easily satisfied by high- κ materials for a wide field region.⁷

Note that it is the quantity ΔG that determines the thermodynamic properties associated with the mixed state of a type-II superconductor.²³ Equation (5) shows that the dependence upon **H** of ΔG is a function only of $h = H/H_{c2}(\theta, \phi)$; in particular, the angular dependence is determined by that of H_{c2} . From Eq. (5) **M**, *S*, and *C* can be derived by $\mathbf{M} = -(\partial \Delta G/\partial \mathbf{H})_T$, $\Delta S = -(\partial \Delta G/\partial T)_{\mathbf{H}}$, and $\Delta C/T = (\partial \Delta S/\partial T)_{\mathbf{H}} = -(\partial^2 \Delta G/\partial T^2)_{\mathbf{H}}$, where $\Delta S = S - S_n$, $\Delta C = C - C_n$, S_n and C_n are the normal-state entropy density and specific heat, respectively. We find

$$-4\pi \mathbf{M} = (H_c^2/H_{c2})\Phi(h)[\hat{H} - (\hat{\theta}/H_{c2})(\partial H_{c2}/\partial \theta) -(\hat{\phi}/H_{c2}\sin\theta)(\partial H_{c2}/\partial \phi)], (7)$$

$$\Delta S(\mathbf{H}, T) = \Delta S(h, T), \tag{8}$$

$$\Delta C(\mathbf{H}, T) = \Delta C(h, T), \tag{9}$$

where $\Phi(h) = -(1/2)dg(h)/dh$. We see that ΔS and ΔC have the same scaling property as that of ΔG , while **M** has additional angular dependence. These scaling properties are useful for deriving relations between the magnitudes of these quantities for **H** along any two different directions; particularly, if the two directions are those parallel to the *c* axis and the *ab* plane of HTSC, we have

$$M_{\parallel c}(H) = \gamma M_{\parallel ab}(\gamma H), \tag{10}$$

$$\Delta S_{\parallel c}(H) = \Delta S_{\parallel ab}(\gamma H), \tag{11}$$

$$\Delta C_{\parallel c}(H) = \Delta C_{\parallel ab}(\gamma H), \tag{12}$$

where $A_{\parallel i}$ $(A = M, \Delta S, \Delta C)$ is the magnitude of the quantity A for $\mathbf{H} \parallel i$ and $\gamma = H_{c2\parallel ab}/H_{c2\parallel c} =$ $(m_3/m_1)^{1/2} > 1$ is the anisotropy ratio; here we have assumed $m_1 = m_2 = m_{ab} < m_3 = m_c$ for HTSC. Note that the component of M transverse to H vanishes when H is along one of the principal axes.⁵ Equation (10) shows, for example, that M for H = 1 T and $\mathbf{H} \parallel c$ is γ times that for $H = \gamma$ T and $\mathbf{H} \parallel ab$. Equations (11) and (12) show that ΔS or ΔC for H = 1 T and $\mathbf{H} \parallel c$ is the same as that for $H = \gamma$ T and $\mathbf{H} \parallel ab$. According to the analysis given in the following, the values of γ ranges from 5 to 8 for YBa₂Cu₃O₇ (YBCO), around 10 for (La_xSr_{1-x})₂CuO₄ (LSCO), and from 15 to 40 for Bi₂Sr₂CaCu₂O₈ (Bi 2:2:1:2).

Note that our theoretical derivation of the scaling relations is done in the mean-field regime, while many ex-

periments have included the region in the H-T plane where fluctuation effects may be important. For the superconducting fluctuation state, the relevant quantity is the coherence energy $\varepsilon = (H_c^2/8\pi)V_c$. For $\mathbf{H} = 0$, the coherence volume $V_c = \xi_{ab}^2 \xi_c$, where ξ_i is the coherence length along the *i* axis; for \mathbf{H} sufficiently large that only the lowest Landau level dominates, $V_c = a_L^2 \tilde{\xi}$, where $a_L = (\phi_0/2\pi H)^{1/2}$ (ϕ_0 is the flux quantum) is the radius of the Landau orbit and $\tilde{\xi}$ is the effective coherence length along the direction parallel to \mathbf{H}^{25} We have $\xi = \xi_c$ for $\mathbf{H} \| c$ and $\xi = \xi_{ab}$ for $\mathbf{H} \| ab$; for the case that **H** does not lie along a principal axis, $\tilde{\xi} = \xi / \sqrt{m(\theta)}$ $[\text{where}\,\underline{\xi=(\xi_{ab}^2\xi_c)^{1/3}}]\,\text{for}\,\kappa^2b\gg 1.^7\,\,\text{We see that}\,V_c(\mathbf{H})\propto$ $1/H\sqrt{m(\theta)}$. Since $\varepsilon \propto (\Delta T)^{3/2}/H\sqrt{m(\theta)}$, the transition width is determined $\underline{by a} \mathbf{H}$ -dependent quantity that is proportional to $[H_{\sqrt{m(\theta)}}]^{2/3}$ (more detailed arguments can be found, for example, in Refs. 25–27). Note that $H\sqrt{m(\theta)}$ has the same angular dependence as that of h [since $H_{c2}(\theta) \propto 1/\sqrt{m(\theta)}$]; therefore, we expect our scaling relations to be valid also in the fluctuation region. The following comparison of these scaling relations with existing experimental data supports our conclusion.

The scaling property of **M** [Eqs. (7) and (10)] has been realized previously in Ref. 28, where the theory has been compared with the reversible magnetization measurements on a Bi 2:2:1:2 single crystal ($T_c \simeq 85$ K) and $\gamma \simeq 18$ as a lower bound (for T = 60 and 64 K) has been found (the uncertainty in γ is due to possible angle misalignment).

We compare the scaling property of C [Eqs. (9) and (12)] with the measurements of Inderhees *et al.*^{8,9} on an untwinned YBa₂Cu₃O_{7- δ} (YBCO) single crystal. From Fig. 2 of Ref. 9, which shows the C/T versus T data for both **H**||*c* and **H**||*ab* and H up to 7 T, we estimate roughly $\gamma \simeq 6-7$ by applying Eq. (12). For example, the curve for H = 1 T and **H**||*ab*. This result $\gamma \sim 6-7$ is close to the result $\gamma \simeq 7.7$ of Farrell *et al.*²⁹ obtained from torque measurements and $\gamma \simeq 7.4$ of Welp *et al.*³⁰ obtained from magnetization measurements for YBCO single crystals, and also is close to that of $\gamma \simeq 5.5$ of the decoration experiments.^{31,32}

Next we point out that the same scaling behavior as that of ΔG , ΔS , and ΔC [Eqs. (5), (8), and (9)] also can be observed in the magnetoresistivity ρ data of HTSC at low values of J (**J** is the transport current density). Generally, we expect that the **H** dependence of ρ is governed by (a) the Lorentz force $(\mathbf{J} \times \mathbf{B})$ and (b) the effect of measuring scale [i.e., H is scaled by $H_{c2}(\theta)$ or $1/\sqrt{m(\theta)}$]. Resistive transition measurements (for example, Refs. 10–19) have shown that ρ depends upon the orientations of \mathbf{J} and \mathbf{H} with respect to the principal axes of the crystal, but is practically independent of the relative orientation of J and H (i.e., the effects of the Lorentz force is practically not observed for both $\mathbf{J} \| ab$ and $\mathbf{J} \| c$; the resistive transition width is found to depend only upon H. Therefore we expect that the angular dependence of ρ of HTSC is mainly governed by the effect of measuring scale of H; i.e.,

$$\rho = \rho(h, T, \mathbf{J}),\tag{13}$$

which gives the relation that for fixed \mathbf{J} ,

$$\rho_{\parallel c}(H) = \rho_{\parallel ab}(\gamma H), \tag{14}$$

where $\rho_{\parallel i}$ is the value of ρ for $\mathbf{H} \parallel i$. Equation (14) means, for example, that ρ for H = 1 T and $\mathbf{H} \parallel c$ is the same as that for $H = \gamma$ T and $\mathbf{H} \parallel ab$. Equation (13) or (14) turns out to be in agreement with the experimental results.^{10–19} In the following we list several examples.

Ive et al.^{10,11} measured ρ versus T at fixed H and ρ versus H at fixed T on a YBCO single crystal for both $\mathbf{H} \| c$ and $\mathbf{H} \| ab$ and H up to 9 T with $\mathbf{J} \| ab$. Their ρ versus T data (Fig. 2 of Ref. 10) satisfy Eq. (14) with $\gamma \sim 6$; for example, the $\rho(T)$ curve for H = 1 T and $\mathbf{H} \| c$ looks almost identical to the one for H = 6 T and $\mathbf{H} || ab$. The fields $H^*(\theta)$, using several different definitions: taking the points $\rho/\rho_n = 0, 10\%, \ldots, 90\%$ (ρ_n is the normal state resistivity), show the same angular dependences as that of H_{c2} (Fig. 8 of Ref. 11) and a $\gamma \simeq 5$ has been estimated by the authors.¹¹ That H^* has the same angular dependence as that of H_{c2} is consistent with our hypothesis [Eq. (13)], which predicts $H^*(\theta) \propto H_{c2}(\theta)$ at a given T. The agreement of the numbers $\gamma \simeq 5$ and 6 with the previous result $\gamma \simeq 6-8$ obtained from C and M measurements and with that $\gamma \simeq 5.5$ of the decoration experiments^{31,32} supports our scaling hypothesis [Eq. (13)].

Kitazawa et al.^{12,13} measured ρ versus T on a large LSCO single crystal for both $\mathbf{H} \| c$ and $\mathbf{H} \| ab$ and H up to 5 T with both $\mathbf{J} \| c$ and $\mathbf{J} \| ab$. Their data (Figs. 1–4 of Ref. 12) satisfy Eq. (14) with $\gamma \simeq 10$. For example, for $\mathbf{J} \| c$ the $\rho(T)$ curve for H = 0.5 T and $\mathbf{H} \| c$ is almost identical to the one for H = 5 T and $\mathbf{H} \| ab$; the same can be observed for $\mathbf{J} \| ab$.

Raffy et al.¹⁷ measured ρ at constant T for H up to 20 T on a c-axis-oriented 1000-Å-thick Bi 2:2:1:2 thin film according to two types of scans: (a) at fixed θ as a function of H and (b) at fixed H as a function of θ . All the curves were reduced to a single one for $0^{\circ} \leq \theta \lesssim 86^{\circ}$ by plotting ρ versus $H\cos\theta$ (corresponding to a scaling factor $1/\cos\theta$). This scaling was based on the idea of Kes et $al.^{33}$ that the dissipation is only related to the transverse field component along the c axis. However, the result of Raffy et al. is also consistent with our scaling hypothesis [Eq. (13)]. The reason is as follows. We believe that the mechanism of superconductivity for HTSC such as YBCO, LSCO, and Bi 2:2:1:2 are the same; therefore their properties should be explainable qualitatively using a single theory. Quantitative differences may exist, such as the magnitude of anisotropy. The scaling using $1/\cos\theta$ as the scaling factor cannot completely explain what has been observed for YBCO and LSCO; therefore, we believe that the $1/\cos\theta$ scaling factor is an approximation valid for large anisotropy, provided that H is not very close to being parallel to ab. Note that $h = H/H_{c2}(\theta) \propto H[\cos^2\theta + (1/\gamma^2)\sin^2\theta]^{1/2} \simeq H\cos\theta$ for $\gamma^2 \gg 1$, provided that θ is not very close to $\pi/2$. Indeed, better fits were obtained including also the region of θ near $\pi/2$ by using $H_{c2}(\theta)$ as the scaling factor (see Fig. 4 of Ref. 17), and $\gamma = 29$ and 36 were found at T = 80 and 83.5 K, respectively. Since the ratio ξ_c/c (c is the lattice constant in the c direction) is

small for Bi 2:2:1:2 (except extremely close to T_c), quasitwo-dimensionality (2D) may be important.^{18,19} As can be seen from Fig. 4 of Ref. 17, $H_{c2}(\theta)$ of a Bi 2:2:1:2 sample is better described by that typical for quasi-2D layered superconductors. Fastampa et al.¹⁹ have recently measured the field H^* (defined as the onset of ρ/ρ_n versus H at fixed T) for a highly *c*-axis-oriented epitaxial Bi 2:2:1:2 film, and shown that the angular dependence of H^* is completely analogous to that of $H_{c2}(\theta)$ and exhibits a crossover from a quasi-2D to -3D behavior near T_c . As mentioned previously, the observation that H^* has the same angular dependence as that of H_{c2} is consistent with the scaling hypothesis [Eq. (13)]. The results of Raffy *et al.*¹⁷ and Fastampa *et al.*¹⁹ thus suggest that our scaling hypothesis holds not only for the cases that H_{c2} follows the angular dependence given by the 3D effective mass model, but also for the case that H_{c2} shows an angular dependence typical for quasi-2D layered superconductors.

The existence of the scaling property for ρ means that the apparently large difference between the magnitudes of the broadening of the resistive transition for $\mathbf{H} \| c$ and $\mathbf{H} \| ab$ is simply due to the large difference in the values of H_{c2} along the two directions, and suggests that the dissipation in the resistive transition region is governed mainly by intrinsic factors. This has an important implication on our understanding of the thermodynamic and electrodynamic behaviors of HTSC. From this result we expect that the effect of the measuring scale of H should also be observed for other quantities such as j_c and T_m . We expect that, to lowest approximation, the scaling behaviors for j_c and T_m are the same as that for ρ , i.e.,

$$j_c(\mathbf{H}, T) = j_c(h, T), \tag{15}$$

$$T_m(\mathbf{H}) = T_m(h). \tag{16}$$

Evidence supporting these scaling hypotheses can be found in the literature. For example, the $j_c(\mathbf{H}, T)$ measurements of Raffy *et al.*²⁰ and Schmitt *et al.*²¹ on Bi 2:2:1:2 films can be accounted for using Eq. (15) including the orientations of **H** near parallel to *ab*. That $j_c(\mathbf{H}||ab, T)$ at fixed *T* is almost *H* independent^{20,21} is simply due to the large values of $H_{c2||ab}$, since a field in the range H = 1 - 10 T corresponds to a very small value of *h* for $\mathbf{H}||ab$ and *T* not very close to T_c . The recent T_m measurements of Beck *et al.*²² on an untwinned YBCO single crystal can also be accounted for using Eq. (16).

The recent ρ measurements of Iye *et al.*³⁴ on both YBCO and Bi 2:2:1:2 samples at higher values of J have revealed the existence of an anomalous peak in $\rho(\theta)$ near the *ab* plane for both compounds; the peak is located much closer to the *ab* plane for Bi 2:2:1:2. The origin of this anomalous behavior is not clear at present. Therefore, the scaling hypothesis of Eq. (13) evidently is valid only for low values of J.

At this point we compare our work with and comment on the theoretical approach of Ref. 35. We mention the following two points. First, the starting points of the angular scaling arguments of both the present work and that of Ref. 35 are the idea of Ref. 4 that F of an anisotropic superconductor in the mixed state can be written in isotropic-like form by a transformation of co-

ordinates (which involves a length rescaling and a rotation). Work along this line has been done previously. 4^{-7} The conclusion for high- κ materials is, as summarized in this paper, that the quantity f in Eq. (1) is a function only of b as shown by Eq. (2), and therefore one has Eq. (5). Equation (5) is the same as its isotropic counterpart except that $\tilde{\kappa}$ or H_{c2} (which serves as the measuring scale for H) is angle dependent; this is the essence of all the scaling arguments of this paper. The results of Ref. 35 [Eq. (4) of Ref. 35] are different from ours since they conclude that for the isotropic-to-anisotropic mapping, in addition to the scaling rule $H \to H \sqrt{m(\theta)} \propto h$, the temperature T should also be rescaled as $T \to \gamma T$; we believe this conclusion is incorrect. Both the energy density and the temperature are coordinate independent; therefore, T, an independent variable, has nothing to do with the transformation. This can be easily seen from the fact that the GL free-energy functional does not involve T explicitly when written in dimensionless form;¹⁻³ the T-dependent GL units $H_c^2/4\pi$ for energy and $\sqrt{2}H_c$ for fields, for example, have nothing to do with anisotropy. Furthermore, Eq. (4) of Ref. 35 is incorrect for M even if one considers the H dependence only; this is because (a) M has additional angular dependences that are not contained in h [see Eq. (7)] and (b) M has a transverse component, which has no isotropic counterpart. Second, it is well known that for conventional isotropic superconductors ρ does depend on Lorentz force $\mathbf{J} \times \mathbf{B}$. The transformation of coordinates used previously⁴ and in Ref. 35 has nothing to do with the disappearance of the effect of Lorentz force in HTSC, since it is only a mathematical procedure for rewriting the anisotropic expression of F in an isotropic-like form and has no physical content. We believe that a complete theory for the behavior of $\rho(\mathbf{H}, T, \mathbf{J})$ of HTSC is not available at present. The argument of Ref. 35, for example, cannot explain the dependence of ρ upon **H** if **J** is parallel to **B** and **J** is along the c axis.

In summary, we have shown theoretically using the GL

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theory in the first half of this paper that the field dependence of ΔG of a high- κ anisotropic type-II superconductor is a function of $h = H/H_{c2}(\theta, \phi)$ for $H \gg H_{c1}$. This property, which we call the scaling property, also exists for other thermodynamic quantities such as $\mathbf{M}, \Delta S$. and ΔC (except that **M** has additional angular dependence). We have argued that these scaling behaviors also hold in the superconducting fluctuation region. We have compared the theory with available experimental results on HTSC, and general agreement has been obtained. In the second half of this paper, based on a careful analysis of existing experimental results, we have pointed out that the same scaling property apparently also exists for ρ , j_c , and T_m of HTSC. The lack of the dependence of the resistive transition width upon $\mathbf{J} \times \mathbf{B}$ indicates that models based on flux-line dynamics alone are unlikely to provide an explanation for the dissipation mechanism in HTSC.^{12,13} The fact that ρ and j_c in nonequilibrium states share the same scaling property with quantities in equilibrium states indicates that the dissipation is controlled by intrinsic factors of the materials, and therefore supports the point of view that thermal fluctuation effects in the presence of a magnetic field are important for HTSC over a quite wide temperature region around the transition. Finally we point out that for low temperatures when the effects of flux pinning become important, the scaling relations for ρ and j_c may not hold; this is because that in addition to the effect of the measuring scale [H is scaled by $H_{c2}(\theta)$ or $\tilde{\kappa}(\theta)$], other effects such as that of the Lorentz force also may be important.

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