Kosterlitz-Thouless transition in Tl₂Ba₂CaCu₂O₈ thin films

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Electrical transport properties of c-axis-oriented $Tl_2Ba_2CaCu_2O_8$ thin films have been examined within the context of the Kosterlitz-Thouless (KT) model. The nonlinear current versus voltage and resistivity versus magnetic field characteristics below the KT transition temperature ($T_c = 99.0$ K), together with the exponential inverse-square-root temperature dependence of the resistivity just above T_c , were consistent with each other and with the theory. The parametrization of the resistivity data well above the transition by Aslamazov-Larkin theory was also consistent with that of the KT theory.

I. INTRODUCTION AND THEORY

The Kosterlitz-Thouless model¹ has been applied to describe phase transitions in a number of two-dimensional systems. These include liquid-solid,¹ magnetic,¹ superfluid,¹⁻³ and superconducting transitions. The latter involve a variety of geometries including ultra-thin films,⁴⁻⁸ and wire and tunneling junction arrays.⁹ One of the unique features of the new high-temperature superconductors is the large anisotropy and essentially twodimensional character of their physical properties resulting from the lavered structure of their conductive planes. Single crystals of YBa₂Cu₃O₇ and Bi₂Sr₂CaCu₂O₈ exhibit large anisotropies in their normal-state resistivities, upper critical fields, and critical currents.^{10,11} Recently the Kosterlitz-Thouless (KT) model has been used to describe the transition to zero resistance in bulk, single-crystal samples of both of these materials.^{12,13} In this paper we report the extension of the use of this model to the superconducting transition of oriented Tl₂Ba₂CaCu₂O₈ films. With their c axes oriented perpendicular to the plane, these films exhibit an upper critical-field anisotropy of about 70.¹⁴ The enhancement of their electrical conductivity above T_c by superconducting fluctuations was previously found to be consistent with the two-dimensional (2D) form of the Aslamazov-Larkin theory.¹⁵

The KT transition in two dimensions involves the unbinding of pairs of topological excitations. In the case of superconductors, the topological excitations are vortices, and the bound pairs consist of vortices of opposite helicity, or vortex-antivortex pairs. The interaction energy between the members of a vortex-antivortex pair is a logarithmic function of the separation, which in the absence of current is given by $U(r)=2\pi Kk_BT \ln(r/\xi)+2E_c$. Here E_c is the vortex core energy, r is the separation of vortex pair, K is the renormalized stiffness constant,¹⁶ and ξ is the Ginzburg-Landau coherence length, which would be the *a-b* plane coherence length in this instance.

Since high-temperature superconducting films which

are several unit cells thick consist of many conducting layers, and are not actually two-dimensional systems, a comprehensive theory must treat the interlayer coupling in some appropriate way. Currently, there is no detailed analysis of this coupling strictly applicable to superconductors. However, in a theoretical study of layered spin systems, which did not treat the magnetic coupling between layers, Hikami and Tsuneto¹⁷ showed that an independent vortex pair configuration in each layer can be energetically favorable relative to a multilayer vortex ring configuration. This situation occurs when the vortex pair separation is less than a critical distance $r_0 = \xi (K/K_c)^{1/2}$, where K_c characterizes the interlayer coupling which for high- T_c superconductors would be in the direction of the c axis. This condition is equivalent to the length scale characterizing the vortex pairs, $l = \ln(r/\xi)$, being less than a critical length scale $l_0 = \ln(r_0/\xi) = \frac{1}{2}\ln(K/K_c)$. It will be shown later that $l_0 \approx 5.3$ near T_c , and that over the entire experimental range all other length scales in the problem are smaller. As a consequence, if magnetic effects between layers are unimportant, there is a rationale for the application of the 2D theory.

The 2D renormalized theory of logarithmically interacting vortices exhibits several key features. As the temperature approaches T_c from below, the average separation of vortices of a pair, ξ_- , diverges and free vortices start to appear at T_c . Near T_c , the corresponding length scale characterizing the renormalized interaction, $l_- = \ln(\xi_-/\xi)$, is given by¹⁶

$$l_{-} \approx \frac{1}{2\pi} \left[\frac{b}{1 - T/T_c} \right]^{1/2}, \qquad (1)$$

where b is constant of order unity. Over the temperature range $T_c < T < T_{c0}$ the average distance between thermally induced free vortices ξ_+ including the effect of the temperature dependence of superelectron density n_s is given as^{6,8,18}

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$$\xi_{+} = a\xi(T) \exp\left[b' \frac{T_{c0} - T}{T - T_{c}}\right]^{1/2}, \qquad (2)$$

where a and b' are constants of order unity. Then the effective density of free vortices $n_f \sim 1/\xi_+^2$ results in a flux-flow resistance proportional to n_f ,⁶ which in the low-current limit is given by

$$R = R_N 2\pi \xi^2 n_f$$

= $A R_N \exp\{-2[b'(T_{c0} - T)/(T - T_c)]^{1/2}\},$ (3)

where R_N is the normal state resistance and A is a constant of order unity.

When $T < T_c$ an applied current will exert a Lorentz force on a vortex pair tending to break it apart. The interaction potential is no longer logarithmic, but possesses a saddle as a consequence of this Lorentz force. The bound vortices can then become unbound by a thermally activated escape process across this saddle in the potential, and the resultant free vortices contribute to the fluxflow resistance.⁸ In the absence of flux pinning this process will result in a nonlinear resistivity proportional to $(J/J_0)^{\pi K}$. The current-voltage characteristic will be of the form $V \sim I^{1+\pi K}$ in the small current limit,^{6,8} where $J_0 = \hbar n_s e / 2m\xi$ is 2.6 times the Ginzburg-Landau critical current density. The length scale associated with this current-induced breaking of vortex pairs is $l_i = \ln(J_0/J)$. An applied magnetic field will also induce free vortices with length scale $l_H = \frac{1}{2} \ln(\eta H_{c2}/H)$,¹² where η is constant of order unity. The other relevant length scales are $l_w = \ln(w/\xi)$ and $l_A = \ln(\lambda^2/d\xi)$ associated with width w of the sample and transverse penetration depth λ^2/d , respectively. Here d will be treated as an effective thickness of an electronic layer of a high- T_c superconductor, whereas it was the film thickness itself in work on low- T_c films. The short *ab*-plane coherence length¹⁴ [$\xi(0) \approx 30$ Å] and effective thickness of an electronic layer¹⁵ ($d \approx 30$ Å) together with $\lambda(0) \approx 0.2 \ \mu m$ (Ref. 12) imply that l_w and l_{Λ} are the order 10, larger than the estimated value of l_0 over the temperature range measurements were performed. It is necessary that l_w be large in order to have a logarithmic interaction between vortices. Finite-size effects are avoided by having l_{Λ} large.

II. SAMPLE PREPARATION

Films of Tl₂Ba₂CaCu₂O₈ were deposited onto ZrO_2-9 mol %Y₂O₃ substrates using a three-gun dc sputtering system equipped with Tl, Cu, and Ba/Ca composite targets. Post-deposition annealing treatments in flowing oxygen with the film and a sample of Tl compound wrapped together in Au foil were necessary to produce high quality films. The x-ray-diffraction pattern showed that the films were single phase with its c axis oriented perpendicular to the plane. The films were not single crystals. The dimensions of the samples used in this measurement were 1.1 cm×0.2 cm×0.7 μ m. Standard dc four-probe methods were used and silver print coating evaporated gold electrodes were used to make contact. All electrical measurements were carried out in

a double-wall μ metal shielded liquid-nitrogen dewar. A solenoid could provide magnetic fields with values up to 280 G and the stray magnetic field inside the shields was <0.02 G.

III. EXPERIMENTAL DATA AND RESULTS

A. The resistive transition

The value of the mean-field transition temperature T_{c0} , used to parameterize Eq. (3) was 100.20 ± 0.3 K. It was determined by fitting the two-dimensional Aslamazov-Larkin fluctuation conductivity,¹⁹ $\sigma - \sigma_N = (e^2/e^2)$ $16h^2d$ $(T/T_{c0}-1)^{-1}$, to the data above T_{c0} . From this fit, the effective thickness d was found to be 34 ± 3 Å.¹⁵ The temperature at which $d^2R/dT^2=0$, 100.5 K, was within the uncertainty in the value of T_{c0} determined from the fit to the Aslamazov-Larkin theory. The result of the fit is shown in the inset of Fig. 1 where the sold line is the fit and circles are the data. The values of $\ln(R_N/R)$ between 99.3 and 99.9 K are plotted as a function of $[(T_{c0}-T)/(T-T_c)]^{1/2}$ in Fig. 1. The best fit to Eq. (3) was found with the parameters $T_c = 98.99 \pm 0.02$ K, $b'=4.30\pm0.02$, $A=1.16\pm0.09$, and $(1-T_c/T_{c0})$ =0.012.

B. Current-voltage characteristics

Current-voltage (I-V) characteristics at various temperatures are shown in Fig. 2(a). The relation $V \sim I^{1+\pi K}$ in the low-current limit can be used to determine the stiffness constant πK from the slope of the plot of $\log_{10} V$ vs $\log_{10} I$. (The range of temperatures over which data could be taken was limited by the fact that transport currents through the finite contact resistances resulted in thermal runaway at temperatures below T=98.72 K.) The values of πK determined from Fig. 2(a) are shown as



FIG. 1. Temperature dependence of resistance is shown in $\ln(R_N/R)$ vs $[(T_{c0}-T)/(T-T_c)]^{1/2}$ plot. The solid line is the fit with $T_c = 98.99$ K over the temperature range 99.3 K < T < 99.9 K. The inset shows the fit of the fluctuation conductivity with the 2D Aslamazov-Larkin theory.



FIG. 2. (a) *I-V* characteristics for various temperatures, (A) 99.64 K, (B) 99.46 K, (C) 99.37 K, (D) 99.27 K, (E) 99.18 K, (F) 99.09 K, (G) 99.00 K, (H) 98.91 K, (I) 98.81 K, and (J) 98.72 K. (b) Magnetoresistivity data for various temperatures, (A) 100.16 K, (B) 99.64 K, (C) 99.46 K, (D) 99.37 K, (E) 99.27 K, (F) 99.18 K, (G) 99.09 K, (H) 99.00 K, (I) 98.91 K, (J) 98.81 K, (K) 98.63 K, (L) 98.35 K, and (M) 97.88 K.

a function of T in Fig. 3(a). With T_c defined as the temperature at which $\pi K = 2$, the KT transition temperature $T_c = 99.0$ K was determined from Fig. 3(a). It was in excellent agreement with the value obtained from the fit of Eq. (3) to the resistance. Ideally πK should exhibit a universal jump from 0 to 2 at T_c for $l = \infty^2$, but finite l and possibly sample inhomogeneity may smear out a sharp jump.⁸ If we assume that the broadening is due only to the finite $l_0 = \frac{1}{2} \ln(K/K_c)$, an approximate value of l_0 near T_c can be obtained from the equation given by Hikami and Tsuneto valid for temperatures slightly above T_c ,¹⁷

$$\frac{\Delta K}{\Delta T} = -\frac{4}{\pi^2} l_0 \ . \tag{4}$$

From the data of Fig. 3(a) using Eq. (4), a value $l_0 \approx 5.3$ was obtained.

C. Magnetoresistivity

The magnetoresistivities versus temperature are also shown in Fig. 2(b). A uniform magnetic field was applied parallel to the c axis. Martin et al.¹² showed, using a



FIG. 3. (a) Temperature dependence of the renormalized stiffness constant πK obtained from Fig. 2. Open and solid circles correspond to data obtained from I-V characteristics and from $\rho(H)$, respectively. The solid line shows the temperature dependence of the unrenormalized stiffness constant πK_0 . (b) Various length scales involved in the measurements plotted as a function of temperature.

heuristic argument, that data of this sort can also be used to determine K through the relation

$$\pi K = 2 \frac{d(\ln \rho)}{d(\ln H)} .$$
⁽⁵⁾

The stiffness constants πK obtained using this alternate method are also shown in Fig. 3(a). The error bars indicate the ambiguities in determining the slopes in the linear regions of the plots of $\ln \rho$ versus $\ln H$ for small fields. For T < 99.09 K, the magnetoresistivities increased very sharply above the noise level after a certain value of field was applied. This might be the consequence of an abrupt onset of the field-induced vortices. A similar behavior was reported in Ref. 12 in the case of $Bi_2Sr_2CaCu_2O_8$ single crystals. We note that these segments of $\ln \rho$ versus $\ln H$ were not included in the determination of the slopes. Furthermore, the range of magnetic fields over which the slopes were determined was chosen in a manner that l_H was always greater than l_- or l_+ as will be described later in connection with the discussion of Fig. 3(b). The stiffness constants obtained from both the *I-V* and $\rho(H)$ measurements generally agree well. However it should be noted that the values of

 πK from $\rho(H)$ data are generally larger by a small amount in the critical region, i.e., within ± 0.5 K of T_c . The above agreement constitutes an important verification of the analysis of Ref. 12, which is actually not supported by a detailed analytical model as is the case for the vortex unbinding by current. All of the results strongly suggest that the superconducting phase transition or oriented Tl₂Ba₂CaCu₂O₈ thin films is well described by KT theory. The differences between the results of the two analyses in the critical region are possibly a consequence of the fact that there may be an additional contribution to the resistance from the unbinding of vortices resulting from the small, but finite, transport current which is not actually included in the $\rho(H)$ analysis. The measuring currents in the $\rho(H)$ measurements were 30 μ**A**.

D. The vortex dielectric constant near T_c

Outside of the critical region, below but close to T_c , renormalization effects should be small and n_s will appear to extrapolate linearly to zero at the mean-field transition temperature T_{c0} as $n_s \sim 1 - T/T_{c0}$.⁸ Consequently in this region, which is the Ginzburg-Landau regime below T_c , $\pi K(T)$ should have the approximate form

$$\pi K(T) \approx \operatorname{const}\left[1 - \frac{T}{T_{c0}}\right].$$
 (6)

Thus an extrapolation from this "low-temperature" region will also determine T_{c0} . Such a straight line is drawn in Fig. 3(a) and the value of $T_{c0} = 100.2$ K determined from the linear fit agrees very well with the meanfield transition temperature obtained from fitting to the 2D fluctuation conductivity above T_{c0} . The constant in the above equation was found to be 340.

The dielectric constant representing the screening of the interaction between a vortex pair by other vortex pairs of shorter pair separation than that of interest at $T = T_c$ is given by^{1,8}

$$\epsilon_c = \pi K_0(T_c)/2 , \qquad (7)$$

where $K_0(T_c)$ is the unrenormalized stiffness constant at T_c . From Eq. (6), we obtain $\epsilon_c = 2.0$. This value is comparable to the value of 1.2–1.8 reported for indium oxides⁷ and Hg-Xe films⁸ but smaller than the value 4.6 reported for a 500-Å-thick YBa₂Cu₃O₇ thin film.²⁰

E. Length scales

In order to observe the fully renormalized KT transition the length scales associated with l_{-} and the experimental probes, l_H or l_j should satisfy the inequalities $l_{-} < l_j$, $l_H < l_0$. It is important to check the validity of these conditions. Taking b'=4.30, and using Eqs. (1) and (2), we obtain $l_{-}=0.026/(1-T/T_c)^{1/2}$ and $l_{+}=0.16/(T/T_c-1)^{1/2}$. These forms are valid very close to T_c , and are connected with the average separation of bound pairs below T_c and average separation of free vortices above T_c , respectively. The quantities $dH_{c2}/dT = -10^4$ G/K (Ref. 14) and $\eta = 0.3$ are then used to evaluate l_H . For the evaluation of $l_i = \ln(J_0/J)$, the Ginzburg-Landau critical current formula $J_0(T) = J_0(0)(1 - T/T_{c0})^{3/2}$ was used, with $J_0(0) = 5 \times 10^5 \text{ A/cm}^2$. The latter was determined by applying Bean's formula²¹ to magnetization measurements on the films. A plot of the various length scales near T_c is shown in Fig. 3(b). It can be clearly seen that the condition $l_i, l_H \gg l_-$ is satisfied for T < 98.9 K. In addition $l_i, l_H > l_+$ holds for T > 99.1 K for small values of current and magnetic field. As a result of l_{-} being the shortest scale length, the measurements determine the fully renormalized values of πK over almost the entire region of interest except a narrow temperature region very close to T_c .

In this narrow temperature region K becomes a function of either l_i or l_H because of the divergence of l_- . Consequently neither the study of vortex unbinding by currents or magnetic fields can determine the fully renormalized parameters. The magnitudes of the measuring current or the magnetic fields determines the length scales over which the renormalized interaction will be probed. Consequently a different approach to the comparison of experiment and theory is necessary. The analytic solutions of the Nelson-Kosterlitz recursion relations,^{2,3} which are valid sufficiently close to T_c , can be used for this purpose in a manner first demonstrated by Kadin et al.⁸ Measurements of the *I-V* characteristics are used in an analysis based on analytic solutions to the recursion relations. The parameters of the I-V characteristics determined above are used together with the unrenormalized K_0 of Eq. (6) to calculate y(l), the vortex excitation probability, as a function of $l = l_i = \ln(I_0/I)$ near T_c . In terms of y, resistance is given by $R(I) \sim yI^2$. We can therefore write $y \sim V/I^3$. To compare with experimental data, we used following scaling relation:⁸

$$v(l) = [V/V_0(T)] / [I/I_0(T)]^3 .$$
(8)

Here $I_0(T) = I_0(0)(1 - T/T_{c0})^{3/2}$ and $V_0(T) = AI_0(T)R_N$ along with the constant A are adjustable parameters. A fit is then obtained by matching the set of theoretical curves, $\ln y(l)$ versus -l, to plots of data, $\ln (V/I^3)$ versus lnI. The result is shown in Fig. 4 over the temperature range 98.81 K < T < 99.09 K. The solid curves are the analytic solutions and l ranges from 2 to 4. Neglecting the high-current regime on the right-hand side of the data, where there may be thermal effects, the theoretical curves and experimental data are in qualitative agreement. The fit is not satisfactory outside of this regime because the analytic solutions work only when $\pi K \approx 2$. From the fit we determine the critical current at the KT transition, $I_0(T_c) = 5.2$ mA. From these values the critical-current density at T=0 is estimated to be $J_0(0) = 3.2 \times 10^5$ A/cm², which is in good agreement with the results of magnetization measurement. This analysis implies that the experimental data very close to T_c are also in agreement with KT theory, but with the length scale of the renormalization determined by the transport current.



FIG. 4. Plots of $\ln(V/I^3)$ vs $\ln I$ at various temperatures. The solid curves are analytic solutions of the KT recursion relations very close to T_c . Numbers indicate corresponding temperatures in Kelvin.

F. Penetration depths

The universal jump condition can be used to estimate the penetration depth⁴ at T_c through the relation $\lambda^2 = \Phi_0^2 d / 32 \epsilon_c \pi^2 k_B T_c$. It is inappropriate to use the "dirty-limit" form here because the coherence length is actually shorter than the electronic mean free path. Using the value of T_c , and the effective thickness d = 34 Å as found by the fit of the Aslamazov-Larkin theory, the unrenormalized penetration depth can be determined.¹⁵ The zero-temperature limit $\lambda(0)$ was found to be either 900 Å from an extrapolation to T=0 using BCS weakcoupling formula²² or 1300 Å using the empirical formula $\lambda^2(T) = \lambda^2(0) / [1 - (T/T_{c0})^4]$. Since d is obtained from the fit over a temperature range at least 2 K above T_{c0} , the effective thickness at T_c might be even larger if the fluctuating layers are more strongly correlated, i.e., thicker, at low temperatures. If d were larger than 34 Å, the above analysis would lead to $\lambda(0) > 900-1300$ Å. This is the order of the values of $\lambda(0)$ for YBa₂Cu₃O₇ (Ref. 23) for $Bi_2Sr_2CaCu_2O_8$ (Ref. 12) which are 1400 Å and 2000-3000 Å, respectively. The magnitude of the penetration depth of Tl₂Ba₂CaCu₂O₈ is not known to us, but a good guess is that it would be that it is the same order as that of the other high- T_c superconductors. This fact would imply that the effective thickness involved in the 2D fluctuation conductivity might be the same as that involved in KT phase transition.

IV. DISCUSSION

The Aslamazov-Larkin theory is concerned with fluctuations in the amplitude of the order parameter above T_c , and the Kosterlitz-Thouless theory is concerned with phase fluctuations and the establishment of quasi-longrange order in the phase below T_c . To the extent that both models account for the data, the behavior of the films is two dimensional, as the 2D-3D crossover of the Aslamazov-Larkin theory was not observed in this material.¹⁵ The underlying superconducting transition resulting in the establishment of local superconducting order is not excluded from being three dimensional by these observations. Indeed, from recent work on ultra-thin conventional superconducting materials, it may have to be so in order for the superconducting transition to occur at a temperature other than T=0.²⁴

An important issue has to do with the role of disorder in these films. Although they are single phase and oriented with their c axes perpendicular to the plane, they are not single crystal. The relative sharpness of the superconducting transition and the agreement between the KT and mean-field transition temperatures determined in a variety of ways, both above and below T_c , suggest that the disorder does not change the nature of the transition although it may renormalize some of the coefficients which are used to describe it. This is consistent with the heuristic argument of Harris²⁵ as to the irrelevance of disorder in the XY model. It should be noted that an array of superconducting grains which is also 2D will exhibit a Kosterlitz-Thouless transition, with a set of renormalized coefficients. We believe this not to be relevant here although the films do consist of crystallites coupled by weak links or junctions of same sort. The reason for this is that the relationship between T_c and T_{c0} in such a situation would be governed by the macroscopic penetration depth, which includes the grain-to-grain coupling. This number we have determined using kinetic inductance measurements²⁶ at 25 MHz to be the order of 1 μ m rather than the 1000-Å value implied by the "universal jump" condition as discussed above.

It is likely that the broadening of the jump in K is associated with an intrinsic property of the sample, the large, but finite anisotropy of the coupling parameters K and K_c . The relationship between the KT coupling constants, the effective mass anisotropy, and the upper critical field anisotropy is as follows:¹⁷

$$l_0 = \ln \sqrt{K/K_c} = \ln \sqrt{m_{\perp}/m_{\parallel}} = \ln(H_{c2\parallel}/H_{c2\perp}) .$$
(9)

The recent measurement of the upper-critical-field anisotropy of very similar $Tl_2Ba_2CaCu_2O_8$ films by Kang *et al.*¹⁴ show an anisotropy of 70, which corresponds to l_0 =4.25. This agrees fairly well with the value of 5.3 obtained from the slope of the stiffness constant just above T_c , given the roughness of the determination of upper critical fields resistively. This agreement makes it highly probable that the observed broadening is intrinsic.

In summary, we have demonstrated that the superconducting phase transition of $Tl_2Ba_2CaCu_2O_8$ thin films is of the Kosterlitz-Thouless-type through studies of both vortex unbinding by a current and by an applied magnetic field. All previous investigations on both high- and low-temperature superconductors have involved one or the other of these,^{12,13} but not both types of investigations. We also find that there is consistency between the parametrization of the 2D characteristics of fluctuation conductivity above T_{c0} and KT transition below T_{c0} .

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These results further confirm the highly-two-dimensional

character of the macroscopic fluctuations associated with

the superconducting transition of $Tl_2Ba_2CaCu_2O_8$ even in samples which are many unit cells thick, i.e., 7000 Å

thick. The results appear to be consistent with a picture in which there is almost no measurable effect of the cou-

pling between layers. Systems which are geometrically

three dimensional in character can behave as two-

dimensional systems only if the effect of the coupling be-

tween the layers is unimportant. This would be the pre-

diction of the theory of Hikami and Tsuneto¹⁷ as all of

the relevant length scales are less than the critical length

scale $l_0 \approx 5.3$. However the precise relevance of this

theory to the multilayer superconducting case would re-

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