

Dimensionality and Kosterlitz-Thouless Transition in Single Grain Tl-2223 Superconducting Thin Films[†]

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Tl₂Ba₂Ca₂Cu₃O_{10+x} (Tl-2223) superconducting films with microstructure consisted of submillimeter size grains combining with reactive ion etching technique, has enabled us to investigate the transport properties of this material in a more controllable manner. Both conductivity fluctuations based on the Aslamazov-Larkin theory and the Kosterlitz-Thouless transitions were studied to delineate the two dimensional nature of the material. It was found that, depending on the substrate used, the effective thickness of critical fluctuation and the detailed features of the K-T transition were very different. For films deposited on LaAlO₃(100) substrates, the effective thickness of critical fluctuation is about 35 Å compared to a value of 17.5 Å obtained for films deposited on MgO(100) substrates, roughly equal to the c-axis lattice constant and the distance between the trilayer CuO₂ planes, respectively. The effective vortex dielectric constant measuring the correlations between vortex pairs near K-T transition were estimated to be 3.0 and 1.6 for films on LaAlO₃ and on MgO, respectively. Possible mechanisms based on the defect structures are proposed to account for the observed results.

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I. Introduction

Although the ultimate mechanism giving rise to high-temperature superconductivity is still a matter of extensive debate [1], it is generally accepted that the correlations between the conducting carriers both within and between the conducting Cu-O layers may have played an important role [2-4]. Within the framework of this conjecture, the two-dimensional (2D) Kosterlitz-Thouless (K-T) transition [5-8] has been proposed as an important clue for understanding high- T_c superconductivity by providing information about

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interlayer coupling via effects of interlayer excitation of vortex rings on the dissociation of independent vortex pairs within the layers [9,10]. The superconducting transition in amorphous He-Xe thin films [11], $\text{Bi}_2\text{Sr}_2\text{Ca}_1\text{Cu}_2\text{O}_{8+\delta}$ (BSCCO) [12], $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) [13] single crystals were interpreted in term of 2DKT theory. The 2DKT transitions in monolayered [14], artificial multilayered [3,4], as well as thin film [15,16] of high-T, superconductors have also been observed. However, despite of the successes of the XT theory in interpreting the experimental results observed over the wide variety of materials cited above, there are some subtle issues remained to be clarified. For instance, how the quasi-2D superconducting "CuO₂ sheets" couple together to give rise the ultimate three-dimensional (3D) superconducting state remains as a challenging issue. Moreover, why both thin films and single crystals of a same material could manifest the similar typical 2D behaviors on the one hand but displayed dramatical differences in transport properties (e.g. the critical current densities can differ by order of magnitudes) on the other hand is also interesting both from fundamental understanding and from practical application point of views.

The other important issue related to the unique layer structure of the newly discovered perovskite superconducting cuprates is the effects of CuO₂ layer spacing on the quasi-two-dimensional nature of these materials. As has been pointed out by Kim et al. [17], the CuO₂ layer spacing can result in strong effect on the nature and, hence, the dynamics of vortices. We note that, although a wide variety of cuprates in various forms have been found to exhibit evidences of 2D behaviors [12-16], results for Tl₂Ba₂Ca₂Cu₃O_{10+δ} (Tl-2223) are still lacking. Since Tl-2223 not only has relatively higher T_c but also larger CuO₂ layer spacing than most of its counterparts, it will be interesting to study the detailed properties of this material and compare that with other cuprate systems provided that pure phased samples can be obtained. In this paper experimental results toward the above mentioned objectives are reported. The unique microstructure, consisting of submillimeter size superconducting Tl-2223 grains obtained by a novel dc-sputtering + postannealing process [18] not only has ensured us that the specimen are indeed single phased Tl-2223 but also has eliminated possible complications resulted from grain boundaries. Evidences that can be unambiguously attributed to the manifestations of 2D systems are demonstrated. The results, however, also indicate that both the effective thickness of 2D conductivity fluctuations and the KT transition related to the dissociation of the bound vortex pairs are very sensitive to the defect structure of the superconducting grains grown on different substrates.

II. Experiments

The detailed procedures and processing conditions of making Tl-2223 superconducting thin films with distinct submillimeter size grains were described previously [18]. Perhaps the most important factor was to raise the postannealing temperature to 910 °C, which was about 20 ~ 30 °C higher than that used in usual processes.

To pattern bridges, typically 1-10 μm in width and 50-100 μm in length, on selected area of the films for subsequent transport measurements the reactive ion etching (RIE) process was employed. Details of RIE conditions for Tl-based films has been reported previously [19]. In the present study, bridges made on single grains of Tl-2223 grown on both MgO (100) and LaAlO₃ (100) substrates were used.

For transport measurements, the typical four probe configuration was used. It is emphasized here that in the present case even the four contact leads were made within the same grain to avoid possible complications originated from intergranular effects. The temperature was monitored and controlled by two separate thermometers attached nearby the sample. All the measurements were carried out in zero applied field and the critical current was defined by an $1 \mu V$ criterion.

III. Results and discussion

Theoretically, for strict 2D systems, it has been known that traditionally defined long-range order does not exist at finite temperatures. As a result, there could be no phase transitions in 2D systems. However, as pointed out by K-T [5], with the dissociation of the bound pairs of positive and negative topological defects in the order parameter, there could be a new type of phase transition in 2D systems. For the so-called K-T transition to occur, the interaction energy between these topological defect pairs has to be logarithmic in the separation distance, τ [6]. In superconducting thin films the vortex-antivortex pairs are the corresponding topological defects as they change the phases of the superconducting order parameter. Although the interaction energy between two vortices is logarithmic in τ only for separations smaller than the transverse penetration depth λ_{\perp} in dirty superconducting thin films with the film thickness $d < \lambda$ (the London penetration depth), $\lambda_{\perp} \approx 2\lambda^2/d$ can be as large as in the order of a few centimeters. As a result, K-T transitions are observable in many conventional superconducting films and high- T_c superconductors [11-16].

Experimentally, one way of probing this phase transition is to directly monitor the dissociation of the bound vortex pairs. Theoretical calculations [6-8] have predicted two main features arising from the dynamics of these unbound vortex pairs: (i) Under the conditions of zero applied field and small measuring currents the resistance originates from the vortex motion induced phase slippage in order parameter is described by the following expression

$$\frac{R}{R_n} = A \exp \left[-2 \left(b \frac{T_{c0} - T}{T - T_{KT}} \right)^{\frac{1}{2}} \right] \quad (1)$$

Where R_n is the normal state resistance and A and b are sample dependent parameters with b on the order of unity. Thus, by measuring the resistance within a temperature range between the mean field transition temperature T_{c0} and K-T transition temperature T_{KT} , one should be able to delineate the manifestation of K-T type phase transition. (ii) Below T_{KT} , the dissociation of the bound vortex pairs can be induced by the externally applied currents. In this case, the current-voltage characteristics (IVC's) are nonlinear with

$$V \propto I^{1+\pi K_R(T)} \quad (2)$$

Where $K_R(T) = m^* k_B T / \hbar^2 n_s^{2D}$ and n_s^{2D} is the 2D carrier density at T . As $T \rightarrow T_{KT}$ from above, $K_R \rightarrow \frac{2}{\pi}$ and the exponent in Eq. (2) displays a universal jump ($1 \rightarrow 3$), known as the Nelson-Kosterlitz jump. In the following we shall show that indeed both features can be identified in the bridges made within single Tl-2223 superconducting grains.

Before we show the results evidencing the existence of the K-T transition based on the above two assertions, it is always instructive to check the quality of the samples used. As has been mentioned previously in the Experiments section, the samples used in the present study were highly c-axis oriented single crystalline Tl-2223 grains with T_{c0} around 110 K. In addition to that, in Fig. 1 we show the J_c 's of two of the representative bridges used in this study to further demonstrate the quality of the samples. As is evident from the results, the J_c 's at 77 K are all well above 10^6 A/cm² with no evidence of weak-link characteristics. The high J_c values of these single grain bridges are comparable to the typical values obtained in most high quality thin film high-T_c cuprates and are indicative of strong pinning effects. As will be discussed below, such strong pinning effects may have played a prominent role in affecting the vortex dynamics associated with K-T transitions to be described below.

In Fig. 2, the in-plane resistances R_{ab} were plotted as a function of temperature for two bridges made of grains grown on different substrates. As can be seen from the results, although the detailed behaviors are somewhat different in the two samples, the basic features predicted by Eq. (1) are unambiguously evident. To further confirm that these are indeed the manifestations of a typical K-T like transition, the corresponding IVC's of the same samples are displayed as a function of the reduced temperature in Fig. 3. Again, the expected Nelson-Kosterlitz jump is observed in both cases. The K-T transition temperatures (T_{KT}) obtained from both kinds of analyses are in very good agreement in both type of films. Namely, with $T_{c0} = 111.3$ K for Tl-2223/MgO, the obtained T_{KT} are 109 K (from R_{ab}) and 108.6 K (from Nelson-Thouless jump), respectively. Whereas for Tl-2223/LAO, with $T_{c0} = 108.2$ K, the corresponding T_{KT} 's are 106.8 K and 106.6 K.

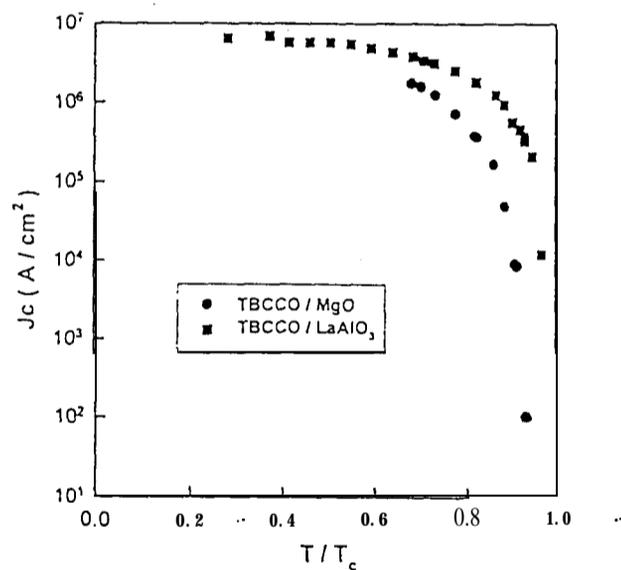


FIG. 1. The critical current density as a function of reduced temperature for bridges made on Tl-2223/MgO (solid circles) and Tl-2223/LaAlO₃ (solid squares).

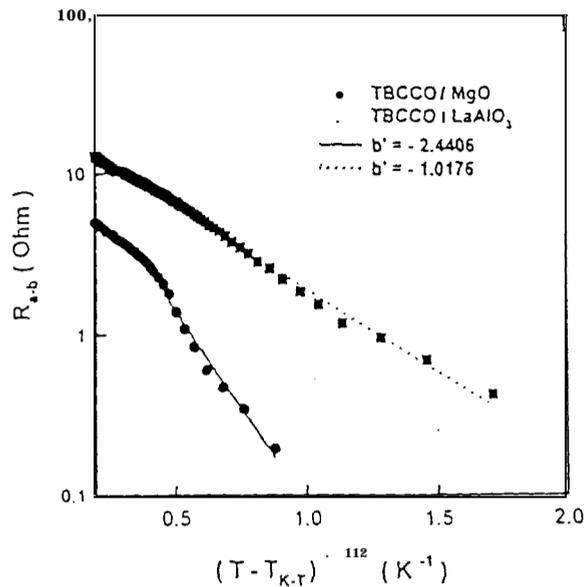


FIG. 2. The semi-logarithmic plot of R_{ab} as a function of $(T - T_{KT})^{-1/2}$, showing that the onset resistances just above T_{KT} follows Eq. (1) closely and is indeed originated from the dynamics of dissociated vortex pairs. The symbols used are the same as those in Fig. 1.

respectively. In any case, all of these results strongly indicated that Tl-2223 is indeed quasi-2D in nature. If we further use the data obtained and taking $\xi_{ab}(0) = 11.3 \text{ \AA}$, $\lambda_{ab}(0) = 2050 \text{ \AA}$ for Tl-2223 [20] to estimate some of the renormalized length scales, we found:

For Tl-2223/MgO, using $\frac{\Delta K}{\Delta T} \approx -\frac{4}{\pi^2} \ell_0$ (Ref. 9), $\ell_w = \ell n(w/\xi)$, and $\ell_\Lambda = \ell n(\lambda^2/d\xi)$, we have $\ell_0 \approx 4.83$, $\ell_w \approx 7.5$, and $\ell_\Lambda \approx 10$, respectively. Here, $\frac{\Delta K}{\Delta T}$, ℓ_0 , ℓ_w , ℓ_Λ , and w are representing the slope of IVC exponent curve near the K-T transition, length scales for vortex pair separations, effective sample dimensions, range for logarithmic interactions, and bridge dimension, respectively. Similarly, for Tl-2223/LaAlO₃, the corresponding length scales are $\ell_0 \approx 4.84$, $\ell_w \approx 8.4$, and $\ell_\Lambda \approx 9.3$, respectively. In each case since $\ell_w, \ell_\Lambda > \ell_0$, and ℓ_0 is larger than other experimental length scales, the system is indeed describable by the 2D theory. We note here that, although the primary features are consistently demonstrated as described above, the origins of the observed differences in grains grown on different substrates remained to be explained.

To resolve this particular issue, we have made further analyses on the obtained data. Since we are dealing with essentially the intrinsic properties of the same material, the most possible origins causing the differences should be the ones that affect the dynamics of the dissociated bound vortices. With this particular clue in mind, we first look into the physical parameter associated with vortex interactions. The physical parameter that describes the screening of the interaction between a specific vortex pair by the presence of other vortex pairs of shorter separation at $T = T_{KT}$ is the vortex dielectric constant ϵ_{KT} . For $T < T_{KT}$, one expects that $\pi K_R(T) \approx \text{const.} [1 - \frac{T}{T_{\infty}}]$ and $\epsilon_{KT} = \frac{\pi K_R(T_{KT})}{2}$. Values of $\epsilon_{KT} \approx 1.6$ and 3.0 were obtained for Tl-2223/MgO and Tl-2223/LaAlO₃, respectively.

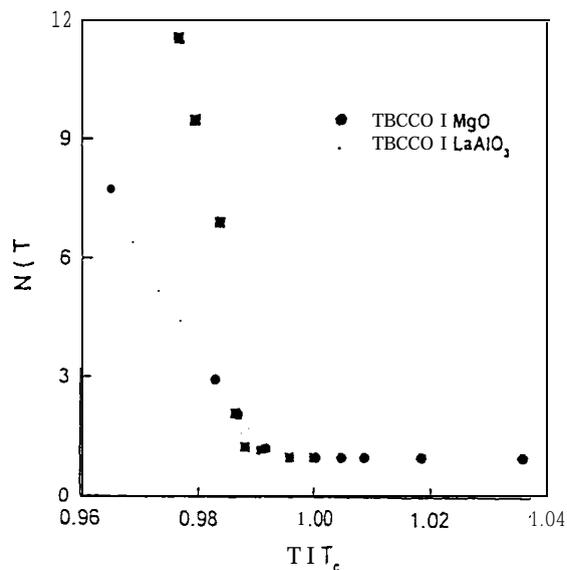


FIG. 3. The Nelson-Kosterlitz jump in the exponent of IVC's in the same bridges.

This, when translating into intuitive physical picture, means that the vortices are more *viscous* in grains grown on LaAlO_3 than that on MgO . Such phenomena may arise either from stronger pinning existent in the CuO_2 layer or from the stronger interlayer correlations between unbound vortices.

Interestingly, when we further analyzed the dimensionality of this material from transport properties above the mean-field transition temperatures the effective thickness in which the 2D behavior displayed similar subtle differences in grains grown on the two different substrates. Fig. 4 shows the results of excess conductivity arising from amplitude fluctuations of order parameter above T_{c0} . As is evident from the results, in bridges made on grains grown on the two different substrates all display the typical 2D behavior over a wide range of temperatures i.e. from T_{c0} to about $2T_{c0}$. The results when fit to the Aslamazov-Larkin expression: [21]

$$\Delta\sigma_{AL} = \frac{e^2}{16\hbar d} \left(\frac{T - T_{c0}}{T_{c0}} \right)^{-1} \quad (3)$$

for 2D superconductors, though all have correct slope of -1, give different value of effective thickness d , with $d = 17.5 \text{ \AA}$ and $d = 35 \text{ \AA}$ for Tl-2223/ MgO and Tl-2223/ LaAlO_3 , respectively. It is noted that 17.5 \AA is about the distance between two trilayered CuO_2 planes and 35 \AA is exactly the c -axis lattice constant of Tl-2223 system. This implies that in Tl-2223/ MgO the conductivity fluctuations take place essentially within the three layers of CuO_2 planes and coupling between groups of the trilayers is negligible. Whereas for Tl-2223/ LaAlO_3 somehow the average coupling between groups of trilayers is stronger. This is in fact quite consistent with the experimental results described above: higher J_c and more pronounced vortex correlations in K - T transition associated effects were evident for bridges made on Tl-2223/ LaAlO_3 . The question is what makes the differences?.

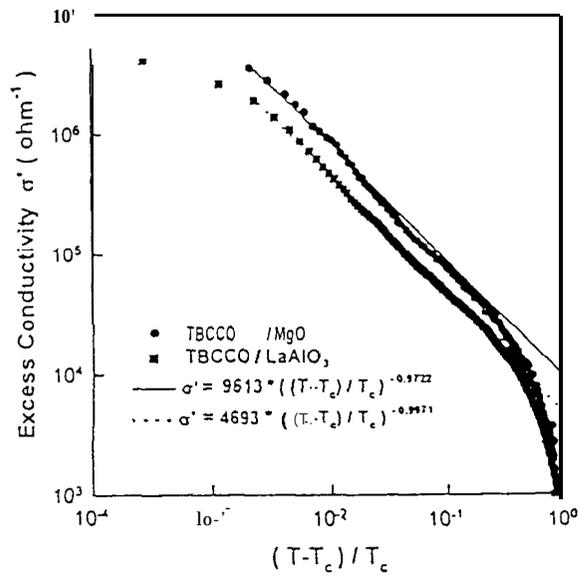


FIG.4. The excess conductivity as a function of reduced temperature for the same bridges shown above. The solid lines are the fits using Eq. (3) with no adjustable parameter except the effective thickness, d .

From the process point of view, the only variation in the two cases is the substrate being different. Since the lattice mismatch is much larger in Tl-2223/MgO ($\approx 9\%$) than in Tl-2223/LaAlO₃ ($< 1\%$), it is expected that the film-substrate correlation during the growth of these submillimeter grains would be much less significant in the former. As a result, the buildup strain energy during grain growth is much smaller in Tl-2223/MgO and the resultant grains become less defective due to the absence of strain relief-induced dislocations. Furthermore, as has been found previously by Kim et al. [22], there was clear evidence that substrate twinning could also result in dislocations in nearby films. Since the crystal structure of LaAlO₃ is cubic only at elevated temperature, above 350-512°C, and become rhombohedral at room temperature, the transition in crystal structure can induce tremendous amount of twins if the process is not slow enough. Thus it is quite natural to expect more defective Tl-2223 grains when grown on LaAlO₃ than on MgO substrates. However, whether this microstructure effect is the sole origin giving rise to the differences or not still have to wait for further detailed investigations. In addition, even if it turns out true, how would crystal defects play such a determinant role in the fundamental properties of these materials should invoke great deal of theoretical understandings.

IV. Summary

In summary, we have demonstrated clear evidences showing that Tl-2223 superconducting phase is indeed quasi-two-dimensional in nature. Parameters derived from vortex dynamics associated with the Kosterlitz-Thouless transitions and the effective thicknesses describing the conductivity fluctuations were given. The implications of the crystalline

defects in affecting the layer coupling and hence the manifestations of order parameter fluctuations, both in phase (i.e. the K-T transition) and in amplitude (i.e. the conductivity fluctuations), are suggested..

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