Vortex unbinding and layer decoupling in epitaxial $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ films

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The current-voltage characteristics of epitaxial Bi₂Sr₂Ca₂Cu₃O_{10+ δ} films in zero applied magnetic field were investigated in a temperature interval of about 20 K below the mean-field critical temperature T_{c0} =110.4 K. Despite the large anisotropy ($\gamma \approx 200$), the data clearly indicate the occurrence of a finite critical-current density, resulting from the Josephson coupling between the (CuO₂)₃ layers. By analyzing the shape of the current-voltage characteristics in terms of quasi-two-dimensional vortex unbinding, it was found that this "intrinsic" critical-current density vanishes well below T_{c0} , just above the hypothetical Kosterlitz-Thouless transition temperature ($T_{\rm KT} \approx 94.8$ K). This is in agreement with Monte Carlo simulations, suggesting a vortex-fluctuation-induced layer decoupling in the case of quasi-twodimensional superconductors. However, close to the layer decoupling temperature, an excess dissipation beyond the quasi-two-dimensional vortex unbinding model appears. This excess dissipation was attributed to the excitation of three-dimensional vortex structures close to the resistive transition.

I. INTRODUCTION

The behavior of thermally excited vortices in layered high-temperature superconductors (HTSC's) has attracted much interest. Although some transport measurements¹ signaled the unique features of the Berezinskii-Kosterlitz-Thouless (BKT) transition,²⁻⁴ corresponding to the pure two-dimensional (2D) case, the presence of Josephson coupling between the superconducting layers should lead to a finite critical-current density.^{5,6}

For a layered superconductor, in the absence of interlayer superconducting coupling, the interaction between the 2D vortices of opposite helicity depends logarithmically on their separation. The vortices and antivortices are bound in pairs below the Kosterlitz-Thouless transition temperature $T_{\rm KT}$. The thermally induced 2D free vortices will be present at $T > T_{\rm KT}$. The creation of free vortices above $T_{\rm KT}$ is thermodynamically favorable due to the entropy contribution to the free energy. $T_{\rm KT}$ is the solution of the equation⁴

$$T_{\rm KT} = \Phi_0^2 s / [8\pi k_B \mu_0 \lambda^2 (T_{\rm KT})] , \qquad (1)$$

where s is the interlayer spacing and λ is the in-plane component of the London penetration depth. Neglecting pinning and size limitation,⁷ the critical current will be essentially zero at all temperatures, the current-voltage (I-V) characteristics having a power-law dependence, $V \sim I^{n(T)}$, with the n(T) variation exhibiting a sudden decrease⁸ from 3 to 1 at $T_{\rm KT}$.

The calculation of the pair energy in the presence of a weak interlayer coupling performed in Ref. 9 revealed another important term in the vortex-antivortex interaction energy, growing linearly with the vortex separation. Following this finding, it was shown in Ref. 6 that the interlayer Josephson coupling gives rise to an "intrinsic" critical-current density J_c decreasing linearly with temperature, at least for T not very close to the mean-field transition temperature T_{c0} , and vanishing at T_{c0} . In such

a situation, no sign of BKT transition should be observed. This can be easily understood in terms of the heuristic argument for a BKT transition.³ With the above linear term in the vortex interaction energy, the free energy of the system with an independent vortex will increase indefinitely with the characteristic system size, irrespective to temperature.

On the other hand, theoretical and numerical studies are in contradiction about the critical behavior near the transition temperature (corresponding to the occurrence of finite resistivity in the small-current limit) in the presence of weak interlayer coupling. Numerical investigations^{10,11} suggest a layer decoupling above the transition. The conclusion that the quasi-2D vortex loop dissociation drives the resistive transition disagrees with theoretical studies 12-14 pointing out that the critical behavior is three dimensional (3D) near the transition, where the correlation length diverges, and with the fact that the renormalized interlayer coupling would increase with each renormalization-group step¹⁵ and there should be no 2D region at all.¹⁶ However, recent reconsiderations of the effect of vortex fluctuations on the interlayer coupling^{17,18} seem to reconcile the Monte Carlo simulation result with that obtained analytically.

The difficulties arising in a BKT description due to the presence of weak interlayer coupling may be avoided, in principle, with some restrictions, in the case of thin films, by considering a BKT transition associated with vortex-string pairing.^{19,20}

In this work, we investigated the change of the I-V curve form with temperature for high-quality $Bi_2Sr_2Ca_2Cu_3O_{10+\delta}$ (Bi-2:2:2:3) films in zero applied magnetic field, down to ≈ 20 K below T_{c0} . The high T_{c0} value of this compound can make the phenomena given by thermal fluctuations more evident. We observed that below ≈ 97 K the sample resistivity determined from the linear part of the I-V characteristics obeys the temperature dependence resulting from a BKT transition, with $T_{KT} \approx 94.8$ K. However, at lower temperatures, the I-V

curves show a clear downward curvature in the log-log plot, indicating the appearance of a finite critical-current density. Furthermore, the approximation $V \sim I^{n(T)}$ for low voltages leads to an n(T) drop larger than 2, and the linear extrapolation of the low-temperature n(T) data gives a T_{c0} value much higher than the real one. The analysis of the *I-V* curve form in the framework of the quasi-2D model from Ref. 6, which attributes the finite critical current to the existence of interlayer Josephson coupling, allowed us to conclude that the interlayer coupling vanishes well below T_{c0} , in agreement with Monte Carlo simulations.^{10,11} The excess dissipation observed when the decoupling temperature is approached from below, beyond the quasi-2D model, can result from the excitation close to the resistive transition of 3D vortex structures (threading many superconducting layers).

II. SAMPLE PREPARATION AND CHARACTERIZATION

Bi-2:2:2:3 films (about 200 nm thick) were prepared by an *in situ* sputtering method on (100) oriented $SrTiO_3$ substrates, as described in detail in Ref. 21. The strongly *c*-axis-oriented growth and in-plane epitaxy were confirmed by x-ray-diffraction studies in a Bragg-Brentano and four-circle geometry. The stacking sequence of the Bi-2:2:2:3 phase was also investigated by transmission electron microscopy, which revealed a low density of stacking faults.

The patterning of the film into a suitable four-probe structure was performed by a standard photolithographic technique, and the electrical contacts were prepared by evaporating and annealing gold. A 2 mm \times 200 μ m strip line was used for transport measurements with pulsed and reversed current. The film investigated in this work has a T_{c0} value of 110.4 K, and the resistivity just above the mean-field transition $\rho(120 \text{ K}) \approx 150 \,\mu\Omega \,\text{cm}$. T_{c0} was determined at the inflection point in the zero-field resistive transition (Fig. 1).

An important parameter involved in the discussion below is the anisotropy factor γ . The γ value can be determined from the crossover field^{22,23} $B_{\rm cr} = \Phi_0 / \gamma^2 s^2$. For applied fields lower than $B_{\rm cr}$, the occurrence of vortex strings with 3D fluctuations is favored, since the intervortex distance becomes larger than the Josephson length $\lambda_J = \gamma s$. It was shown recently²³ that B_{cr} can be obtained from conventional resistive measurements, with the applied magnetic field parallel to the c axis, where a change in the magnetic field dependence of the activation energy in the thermally assisted flux flow regime can be observed. For the sample investigated in this work, a $B_{\rm cr}$ value of 0.1 T was determined,²⁴ and the above relation gives, with the distance between the $(CuO_2)_3$ layers s = 1.86 nm, $\gamma \approx 120$. However, in the case of a large anisotropy $(\gamma > \lambda/s)$, the contribution of the magnetic interaction to the increase of the tilt modulus has to be considered,²⁵ and then $B_{\rm cr} \approx 4\Phi_0 / \gamma^2 s^2$, leading to a γ value two times larger. Below we will use an anisotropy factor of the order of 200.



FIG. 1. The resistive transition of the investigated Bi-2:2:2:3 film in zero applied magnetic field, for a transport-current density of 2.5×10^7 A/m². The mean-field transition temperature (determined at the inflection point) is $T_{c0} = 110.4$ K.

III. RESULTS AND DISCUSSION

The *I-V* curves for the temperature interval between 90 and 98 K are illustrated in Fig. 2. The change of the curvature of the *I-V* curves in the double logarithmic plot resembles phenomena associated with the transition to a vortex glass in zero applied magnetic field.²⁶ For our sample, in the sensitivity window of our measurements, the glass temperature would be $T_g \approx 94.2$ K (linear *I-V* curve in the log-log plot at low levels, Fig. 2). At the glass transition, $V(T_g) \sim I^{(z+1)/(d-1)}$. The exponent $n(T_g) = (z+1)/(d-1) \approx 5.5$ would give a dynamic critical exponent $z \approx 10$, if we consider the dimensionality d=3 (3D vortex structures). This value is considerably higher than that predicted for the transition to the Meissner state (z=2, Ref. 26).

In a very crude approximation, one can take an averaged slope of the I-V curves in Fig. 2, for a limited interval, in order to check the appearance of the Kosterlitz-



FIG. 2. The current-voltage characteristics in zero applied magnetic field (magnetically shielded environment) at different temperatures. From right to left: T = 90 K; 91.2 K; from 92 up to 94 K in steps of 0.4 K; from 94 up to 96.6 K in steps of 0.2 K; 97 K; 98 K.

Nelson drop in the n(T) dependence. The temperature variation of such an exponent, determined between 0.1 and 10 μ V, is represented in Fig. 3. While a precipitous change between ≈ 94 and 96 K is evident, the disagreement with a BKT transition (associated with 2D vortex pairing or vortex-string pairing) is obvious. The "jump" is larger than 2, and the linear n(T) variation observed at low temperatures gives by extrapolation to n = 1 a mean-field transition temperature much larger than the real T_{c0} (see Fig. 3).

Another characteristic behavior, as can be seen in Fig. 2, is that above ≈ 95.8 K, in the low-I limit, the I-V curves are Ohmic. In this temperature range we are dealing with vortex fluctuations, as confirmed in Ref. 7, where it was shown that in the case of Bi-2:2:2:3 samples with preferential grain orientation, in the condition of a strong size limitation, the resistive tail disappears. The size limitation, caused by ribbonlike crystallites and "good" superconducting intergranular contacts with a small extension w in the (a,b) plane, leads to a finite critical-current density $\sim 1/w$, even in the 2D case (i.e., for quasi-2D HTSC's, when $w \ll \lambda_J$). For our sample, the resistivity determined from the I-V curves in the linear region, above but near 95.8 K, depends on temperature similar to what is expected in the case of a BKT transition, with $T_{KT} \approx 94.8$ K (see Fig. 4). Note that due to the large difference between T_{c0} and T_{KT} , the data in Fig. 4 for which a good agreement with the BKT description appears, correspond to $T(T_{c0} - T_{KT})/T_{KT}(T_{c0} - T)$ $-1 \approx 0.1$. Close to $T_{\rm KT}$, the correlation length ξ_+ , representing the scale at which vortices begin to unbind, takes the exponential form²⁷

$$\xi_{+}(T) \approx \xi(T) \exp\{b[(T_{c0} - T)/(T - T_{KT})]^{1/2}\},$$
 (2)

where ξ is the coherence length in the (a, b) plane, and b is a constant of the order unity. With the density of free, unpaired vortices above $T_{\rm KT}$, $n_f \sim 1/\xi_+^2$, the resistivity in the flux flow regime will be

$$\rho(T) \sim \exp\{-2b[(T_{c0} - T)/(T - T_{KT})]^{1/2}\}.$$
 (3)

For our sample, b=2.6. It is worth noting that the above $T_{\rm KT}$ value and Eq. (1) lead with $\lambda(T) = \lambda(0)(1-T/T_{c0})^{-1/2}$ to $\lambda(0)=165$ nm, and, with



FIG. 3. Temperature dependence of the exponent *n*, determined between 0.1 and 10 μ V.



FIG. 4. Temperature variation of the reduced resistivity fitted at low levels with Eq. (3) ($T_{\rm KT}$ =94.8 K and the parameter b=2.6). The resistivity was determined from the linear part of the *I*-V curves in the low-*I* limit (Fig. 2).

 $\lambda(T) = 0.7\lambda(0)(1 - T/T_{c0})^{-1/2}$, to $\lambda(0) \approx 230$ nm. These values are close to the experimental determination²⁸ $[\lambda(0) \approx 200 \text{ nm}].$

Taking into account the above features, one may think that free 2D vortices occur in the system with interlayer coupling when ξ_+ becomes smaller than λ_J , implying a 3D-2D crossover at a temperature²⁹

$$T^* = T_{\rm KT} + (T_{c0} - T_{\rm KT}) / \{1 + (1/b^2) [\ln(\lambda_J / \xi)]^2\}, \quad (4)$$

[see Eq. (2)]. $T_{\rm KT}$ corresponds to the BKT transition which should appear in the system in the absence of interlayer coupling. However, with $\xi(T) = \xi(0)(1 - T/T_{c0})^{-1/2}$ [$\xi(0) = 2 - 3$ nm] and the $T_{\rm KT}$, b, and λ_J values, Eq. (4) gives $T^* = 99.7 \pm 0.4$ K, which is clearly higher than the lowest temperature value at which the flux flow behavior was observed.

The above inconsistencies seem to be closely related to the existence of Josephson coupling between the layers, leading to the occurrence of a finite critical-current density, and this will be discussed below.

Following Refs. 6 and 9, the energy E of a 2D vortex pair of separation r for coupled layers, in the presence of a transport current I flowing parallel to the (a,b) plane, is given by

$$E(r) \approx 2E_c + E_1 \ln(r/\xi) + E_2(r/\xi) - (s\Phi_0/A)Ir , \qquad (5)$$

where E_c is the creation energy, E_1 is the coupling constant in the (a,b) plane, $E_2 = \pi E_1 \xi / \sqrt{2} \lambda_J$, and A is the sample cross section. (For the definition of the in-plane and the out-of-plane coupling constants, see Ref. 6). The second term on the right-hand side of Eq. (5) is the usual logarithmic interaction, the third term results from the presence of interlayer Josephson coupling, and the fourth represents the modification of the interaction energy due to the Lorentz force. With Eq. (5), the analysis of the current-induced vortex-antivortex unbinding⁶ leads to a current-voltage characteristic of the form

$$V \sim I(I - I_c)^a . \tag{6}$$

Here,

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$$a = E_1 / 2k_B T \sim 1 / T - 1 / T_{c0} , \qquad (7)$$

and the critical current I_c is given, up to a constant factor, by the ratio E_2/ξ . When $I >> I_c$, Eq. (6) reduces to the result for 2D superconductors (n = a + 1). The voltage occurs for $I > I_c$ through current-induced flow of unbound, free vortices, created at a relative distance larger than the characteristic intervortex separation for which the pair energy E(r) from Eq. (5) has its maximum, i.e.,

$$r_m = E_1 A / [s \Phi_0(I - I_c)] = 2 A a k_B T / [s \Phi_0(I - I_c)] .$$
 (8)

At low temperatures, in the investigated temperature interval, the I-V curves of our sample fit Eq. (6), in a threeparameter fit, as illustrated in Fig. 5. Close to the resistive transition, however, good fit is obtained only if some data at low voltage levels are left out of consideration. Besides this aspect, which will be discussed further, the resulting exponent exhibits a temperature variation in good agreement with Eq. (7) (see Fig. 6), which, in our opinion, constitutes a strong experimental support for the above simple model. The temperature dependence of the critical current obtained from the fit is shown in Fig. 7. The important point here is that I_c vanishes at around 95.5 K, indicating layer decoupling, as suggested by Monte Carlo simulations.^{10,11} The critical current determined with a low voltage criterion has a similar behavior.

The appearance of an excess dissipation, beyond the current-induced quasi-2D vortex unbinding model, seems to result from the excitation, near the resistive transition (where the correlation length diverges), of 3D vortex structures, which drive the transition. In the case of a 3D vortex structure threading a few (Cu O₂)₃ layers, the form of the *I-V* curve remains practically the same, but the critical current will be reduced.³⁰ As a limiting situation, the free energy of a quasi-2D vortex-antivortex pair of separation $r \gg \lambda_J$ can be higher than the free energy of a vortex-string pair threading the whole film and having the same separation (in the latter case, the interaction energy is free from the Josephson coupling term^{19,20}). The estimations made in Ref. 20, applied in the case of our



FIG. 5. The current-voltage characteristics at different temperatures between 88 K (right) and 94.8 K, in steps of 0.4 K. The continuous lines represent the fit with Eq. (6).



FIG. 6. Temperature dependence of the exponent *a*. The continuous line represents the one-parameter fit with Eq. (7) $[a = \text{const.}(1/T - 1/T_{c0}), \text{ with } T_{c0} = 110.4].$

sample and for an interlayer coupling term linear in r, indicate that, by taking into account the entropy contribution to the free energy due to thermal distorsions,³⁴ the vortex-string excitations become favorable for separations larger than $\approx 20 \,\mu\text{m}$ at T around 92 K. This is the length scale r_m (Ref. 31) resulting from Eq. (8), with I- I_c of the order of 3 mA and $a \approx 3$, for $T \approx 92.4$ K (see Figs. 5–7). Above this temperature value, in the sensitivity window of our measurements, a significant excess dissipation at low transport currents can be observed (Fig. 5).

In a system with decoupled layers, an isolated vortex line threading the layers disintegrates into 2D vortices.^{32-34,29} We determined the activation energy for our sample in the thermally assisted flux flow regime, with the applied field B (2 mT and 2T) parallel to the c axis. The activation energy, U(B,T), was obtained from resistive data, $\rho(B,T)$, assuming that the activation energy vanishes at the mean-field critical temperature T_{c0} ,

$$U(B,T) = T[\ln\rho(B,T_{c0}) - \ln\rho(B,T)]$$
(9)

 $(k_B = 1)$. T_{c0} does not depend significantly on *B*, at least up to 2 T. Due to the limited sensitivity of our experiments, the resistivity was measured at a transport-current



FIG. 7. Temperature dependence of the critical current I_c determined at 0.1 μ V (\bullet) and resulting from the fit of the current-voltage characteristics with Eq. (6) (\odot). I_c vanishes at $T \approx 95.5$ K, indicating layer decoupling.



FIG. 8. The activation energy U determined with Eq. (9) vs temperature, for B=2 mT (right) and B=2 T. The rapid U(T) decrease for B=2 mT at around 95.5 K may be partially associated with the decoupling of the $(CuO_2)_3$ layers above this temperature.

density of 2.5×10^7 A/m². As can be seen in Fig. 8, for B=2 T the activation energy shows a linear temperature decrease, extrapolating to zero close to T_{c0} ,³⁵ whereas, for B=2 mT, a strong U(T) decrease at around 95.5 K appears. Since at this temperature the field value B=2 mT is lower than Φ_0/λ^2 , the vortices can be considered in the isolated limit, and the U(T) drop may be associated with the occurrence of vortex-string disintegration above the layer decoupling temperature.

IV. CONCLUSIONS

The *I-V* characteristics of epitaxial Bi-2:2:2:3 films in zero applied magnetic field, investigated in a temperature interval down to ≈ 20 K below the mean-field critical temperature, indicate the occurrence of a finite critical-current density due to the existing Josephson coupling

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between the $(CuO_2)_3$ layers. By analyzing the shape of the *I-V* curves in the framework of the single-pair, quasi-2D vortex-unbinding model (which seems to be a good approximation, at least for a vortex density lower than $1/\lambda_J^2$), it was shown that the interlayer Josephson coupling vanishes well below the mean-field critical temperature, just above the hypothetical BKT transition temperature, in agreement with the Monte Carlo simulation result. This would be the reason why the resistive data in the case of highly anisotropic HTSC's exhibit a temperature dependence similar to that predicted by the BKT description, in the critical region, or by the 2D Ginzburg-Landau Coulomb gas model outside the critical region. The vanishing of the interlayer coupling means a divergency of the Josephson length. In this situation, the upper 3D-2D crossover (above the resistive transition) is shifted to a temperature value lower than that predicted by Eq. (5).

The excess dissipation, beyond the quasi-2D vortex unbinding model, appearing when the layer decoupling temperature is approached from below, seems to result from the excitation of 3D vortex structures, which can drive the resistive transition through a pinning-assisted unbinding process or a vortex-glass transition.

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value in Eq. (7) would be reduced (Ref. 7) by a factor (a-1)/a. The effect is negligible at low temperatures $(a \gg 1)$.

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