Doping-Induced Change in the Interlayer Transport Mechanism of $Bi_2Sr_2CaCu_2O_{8+\delta}$ near the Superconducting Transition Temperature

S. O. Katterwe, A. Rydh, and V. M. Krasnov*

Department of Physics, Stockholm University, AlbaNova University Center, SE-10691 Stockholm, Sweden

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We perform a detailed study of temperature, bias, and doping dependence of interlayer transport in the layered high temperature superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$. We observe that the shape of interlayer characteristics in underdoped crystals exhibits a remarkable crossover at the superconducting transition temperature: from thermal activation-type above T_c to almost T-independent quantum tunneling-type below T_c. Our data provide insight into the nature of interlayer transport and indicate that its mechanism changes with doping: from the conventional single quasiparticle tunneling in overdoped to a progressively increasing Cooper pair contribution in underdoped crystals.

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How does high temperature superconductivity (HTSC) emerge with decreasing temperature? This highly debated question is crucial for understanding HTSC. The superconducting transition in HTSC is unusual, with a seeming lack of changes in the quasiparticle (QP) density of states at T_c and persistence of the normal state pseudogap above T_c [1,2]. However, there is no consensus about the T evolution of the energy gap. The results range from complete T independence [3] to a strong T dependence of the gap at $T \to T_c$ [4–9]. In some experiments, the coexistence of the two energy scales below T_c was reported [7–10]. The existing controversy is one of the major obstacles for understanding HTSC and requires further T-dependent studies.

Intrinsic tunneling spectroscopy utilizes the weak interlayer (c-axis) coupling in layered HTSC [7,8,11–14]. Unlike surface probe techniques, it is perfectly suited for T-dependent studies and provides information about bulk electronic properties. Furthermore, it gives a possibility to measure the QP resistance at different bias at $T < T_c$ [11,12,15] (which is otherwise shunted by the supercurrent [16]) due to the large specific capacitance of intrinsic junctions [17], allowing junctions to remain in the resistive state even below the critical current. Thus, bias yields an additional parameter for intrinsic tunneling studies, which may render crucial for correct interpretation of the data.

Here we study variation of intrinsic tunneling characteristics in small $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) mesa structures, upon transition through T_c. We observe an abrupt crossover in the shape of current-voltage (I-V) characteristics, from thermal activation (TA) like above T_c to T-independent quantum tunneling like below T_c. Remarkably, the crossover is most clearly seen in underdoped Bi-2212, in which no other spectroscopic features can be resolved at T_c. Our data indicate that the interlayer transport mechanism changes with doping, from single QP tunneling in overdoped to progressively increasing pair tunneling in underdoped Bi-2212. The observed simple TA behavior in the whole normal state region and in a wide bias range actuates the question of whether the c-axis pseudogap represents the pairing gap.

We study two batches of crystals: the Y-doped Bi(Y)-2212 with the maximum $T_c \simeq 94.5$ K and the pure Bi-2212 with the maximum $T_c \simeq 86$ K. Details of mesa fabrication and characterization are described elsewhere [7,8,13].

Figures 1(a) and 1(b) show the T variation of $I - V$ and $dI/dV(V)$ curves for an underdoped Bi-2212 mesa with $N = 7$ intrinsic Josephson junctions; see Fig. 1(c). The following characteristic features are seen in dI/dV curves: The sharp "coherence" peak appears at $T < T_c$ and, in analogy with conventional low-T_c junctions, is attributed

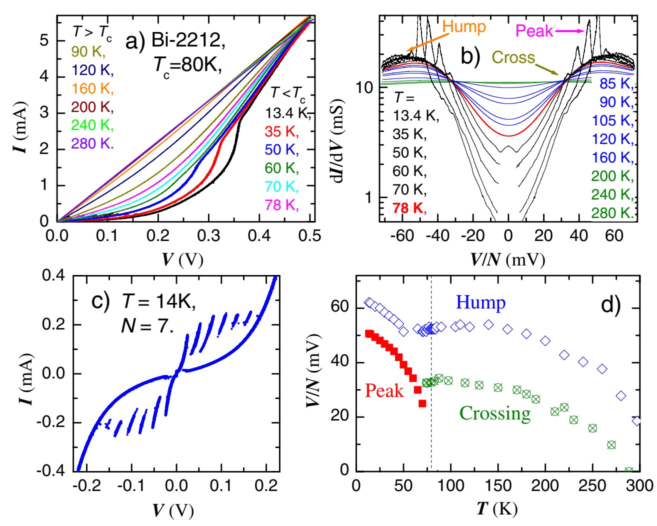

FIG. 1 (color online). (a) I-V and (b) $dI/dV(V)$ characteristics at different T for an underdoped $N = 7$ Bi-2212 mesa with $T_c \simeq 80$ K. (c) Periodic quasiparticle branches caused by sequential switching of intrinsic junctions from the superconducting to the resistive state. The I-V curve is hysteretic; i.e., switching to the resistive state occurs at a larger current than retrapping back to the superconducting state. (d) Temperature dependence of the peak, hump, and crossing voltages (per junction). The vertical dotted line represents the T_c.

to the sum-gap voltage $2\Delta/e$ [7,8,11,14]. A broader hump is seen in the whole T range and is associated with the c-axis pseudogap [7,8,11]. At elevated T, the dI/dV curves cross in nearly one point [8,14]. The T dependence of these features is plotted in Fig. 1(d).

Figure 2(a) shows $dI/dV(V)$ curves (in a semilog scale) for the most underdoped Bi(Y)-2212 mesa with $N = 34$ junctions, both below and above T_c. It is seen that the shape of interlayer characteristics exhibits a remarkable qualitative change at T_c: (i) Above T_c, the average slope of $\ln[dI/dV](V)$ curves decreases with increasing T, while the voltage scale remains almost the same. The curves lean towards the horizontal line, corresponding to the normal state conductance, and cross almost in one point. (ii) Below T_c, the $\ln[dI/dV](V)$ curves have almost *the same T-independent slope*. With increasing T, the curves remain parallel and move towards the vertical axis $V = 0$ (i.e., the voltage scale decreases).

Experimental curves from Fig. 2(a) have a characteristic V shape with the slope of each leg approximately independent of voltage. The curves at $T > T_c$ closely resemble TA characteristics [18]. The TA conductance can be described by a simple expression [19]:

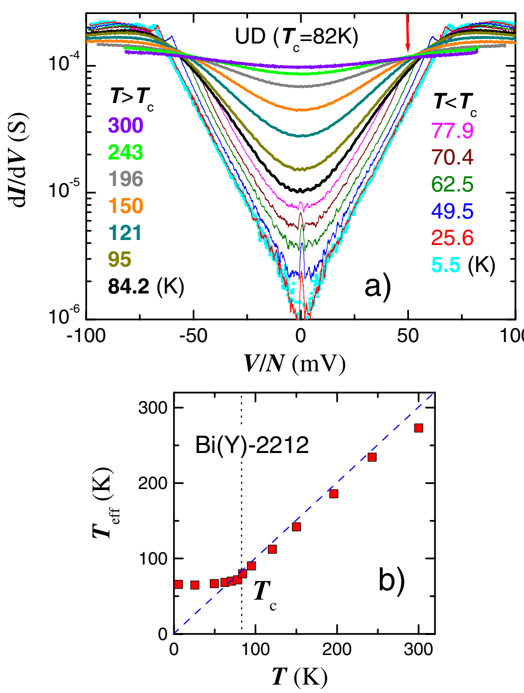

FIG. 2 (color online). (a) A semilog plot of $dI/dV(V)$ curves for an underdoped Bi(Y)-2212 mesa at different T. Note that below T_c the curves maintain the same slope, while above T_c the slope changes progressively with T. A characteristic crossing point of $dI/dV(V)$ curves at $T > T_c$ is marked by the arrow. (b) The effective temperature, obtained from the slopes of the $dI/dV(V)$ curves at a finite bias $V/N = 30$ mV. A remarkable crossover from the thermal activation behavior $T_{\rm eff} = T$ (shown by the dashed line) to the quantum behavior $T_{\rm eff} = $ const occurs at T_c (the vertical dotted line).

$$\frac{dI}{dV}(T, V) \propto \frac{n}{T} \exp\left[-\frac{U_{\rm TA}}{k_B T}\right] \cosh\left[\frac{eV}{2k_B T}\right], \qquad (1)$$

where n is the concentration of mobile charge carriers and $U_{\rm TA}$ is the TA barrier. Indeed, the cosh term reproduces the rounded V shape of $\ln[dI/dV](V)$ curves with the slope that increases as $\simeq 1/T$.

Equation (1) with constant $U_{\rm TA}$ describes very well the data from Fig. 2(a) in the whole normal state $T > T_c$ [19]. It also reproduces the crossing point [18], which according to Eq. (1) occurs at $eV \sim U_{\rm TA}$. Thus, the crossing point is simply a consequence of a fairly T-independent c-axis pseudogap [1,3,7,8], while its voltage represents the pseudogap energy. Indeed, a correlation between the hump and the crossing voltages is seen from Fig. 1(d).

According to Eq. (1), the slope of the curves in Fig. 2(a) should be approximately proportional to $1/T$. Indeed, $d/dV(\ln[dI/dV]) = (e/2k_BT)\tanh(eV/2k_BT) \simeq e/2k_BT$ for $eV > 2k_BT$. Figure 2(b) shows the effective temperature $T_{\rm eff}$ obtained from the slopes at $V/N = 30$ mV, using Eq. (S4) from Ref. [19]. It is seen that in the normal state $T_{\rm eff} \simeq T$, confirming the TA nature of interlayer transport at $T > T_c$. However, at $T < T_c$ an abrupt saturation of $T_{\rm eff}(T)$ occurs, typical for quantum tunneling transport [17,20]. This is the central observation of this work. Remarkably, the crossover is clearly distinguishable at $T \to T_c$, although no other spectroscopic features can be resolved in $dI/dV(V)$ curves.

So far, we have discussed $dI/dV(V)$ at finite bias. Another straightforward way to investigate the TA behav-

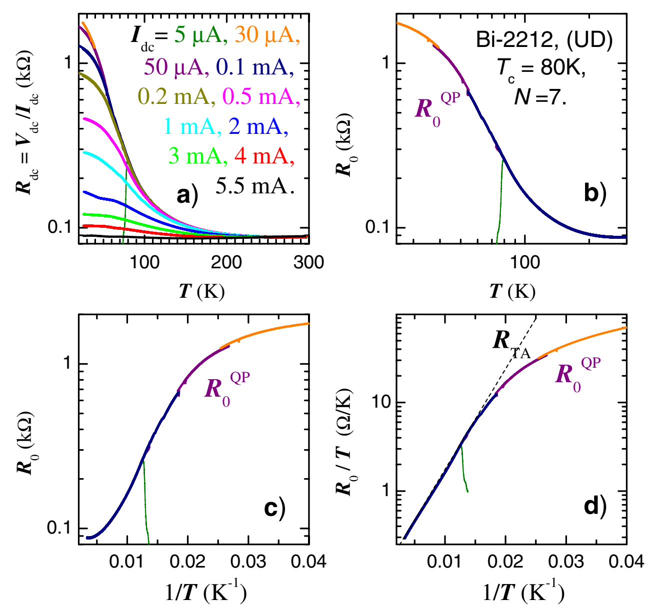

FIG. 3 (color online). (a) T dependence of the dc resistance at different bias currents for the same Bi-2212 mesa. Panels (b)–(d) show the asymptotic zero-bias resistance: (b) in the double-logarithmic scale; (c) as a function of $1/T$; and (d) normalized by T: a clear linear thermal activation behavior (dashed line) is observed in the whole normal state region.

ior is to consider T dependence of zero-bias resistance R_0 [the exp term in Eq. (1)]. Here we benefit from the ability to measure the QP resistance at different bias $R^{QP}(I)$. Figure 3(a) shows the dc resistance $R_{dc} = V/I$ at different I for the same Bi-2212 mesa as in Fig. 1. Measurements were done by first applying a current, sufficient for switching to the last QP branch [see Fig. 1(c)] and then ramping it back to the desired value. The R_{dc} at the smallest current $I = 5\ \mu A$ coincides with the conventional ac resistance R_{ac}. It drops at $T < T_c$, since the finite quality factor of the junctions causes retrapping to the superconducting branch at a finite current [17]. From Fig. 3(a) it is seen how nonlinearity develops with decreasing both I and T. At small I, the curves approach an asymptotic, allowing determination of the zero-bias QP resistance R_0^{QP} without extrapolation.

The zero-bias resistance grows with decreasing T and is usually described either in terms of power-law dependence [16], inherent for single QP tunneling in the presence of a d-wave gap, or in terms of TA [21]. The double-logarithmic plot [Fig. 3(b)] demonstrates that $R_0(T)$ for our mesas is not described by the power law in any extended T range. Neither is it perfectly described by the Arrhenius law $\exp(U_{TA}/k_BT)$, as demonstrated in Fig. 3(c). On the other hand, Fig. 3(d) demonstrates that the ratio R_0/T follows very accurately the Arrhenius law (dashed line) in the whole normal region, consistent with the TA expression Eq. (1) and with the finite bias behavior [Fig. 2(b)]. However, at $T < T_c$, R_0^{QP}/T deviates downwards from the Arrhenius law. This is consistent with saturation of $T_{eff}(T < T_c)$, as shown in Fig. 2(b). Therefore, it is a consequence of the same crossover.

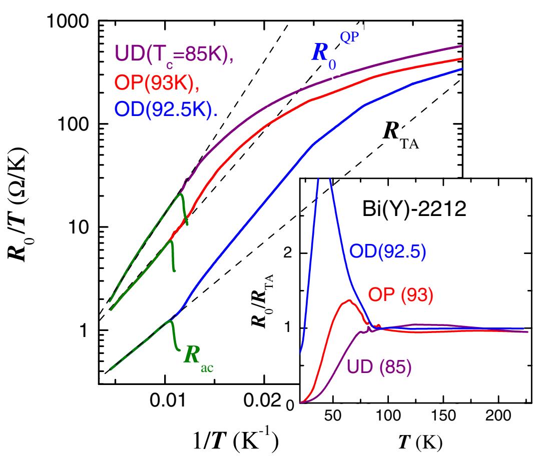

FIG. 4 (color online). T-normalized zero-bias quasiparticle resistance R_0^{QP} extrapolated from I-V curves at $T < T_c$ and R_{ac} measured with a small ac current for Bi(Y)-2212 mesas with different doping: UD, underdoped; OP, optimally doped; OD, overdoped. Dashed lines represent TA fits [Eq. (1)] at $T > T_c$. The inset shows relative values of the excess QP resistance with respect to the TA fit of the normal state. A large excess resistance appears below T_c in the overdoped mesa. The excess resistance rapidly decreases with decreasing doping and becomes negative in the underdoped mesa.

In Fig. 4, we analyze the doping dependence of $R_0(T)/T$. In the normal state, it varies in the TA manner for all studied doping levels. In the superconducting state, R_0^{QP} also continues to grow with decreasing T, but the rate of the growth with respect to the TA behavior R_{TA} (dashed lines) strongly depends on doping:

For the overdoped mesa, $R_0^{QP}(T < T_c)$ grows much faster than in the normal state; i.e., there is a sharp onset of the excess resistance at $T = T_c$. However, the excess resistance at $T < T_c$ decreases rapidly with underdoping. It becomes small in the near optimally doped mesa and turns *negative* already in moderately underdoped mesas.

The inset in Fig. 4 shows the magnitude of the excess QP resistance with respect to the TA fit [a minor max 5% deviation from $R_0/R_{TA} = 1$ at $T > T_c$ in the underdoped mesa can be attributed to a modest growth of $n(T)$ [22]]. A progressive decrease of the excess QP resistance at $T < T_c$ with decreasing doping is obvious.

We start a discussion of the observed phenomena by recollecting the expected T variation for conventional single QP tunneling characteristics [19]: (i) Opening of the superconducting gap at $T < T_c$ leads to appearance of the large excess QP resistance due to rapid freezing out of QPs upon their condensation into Cooper pairs. (ii) R_0^{QP} continues to grow with decreasing T both due to freezing out of thermal QPs and due to growth of the QP lifetime, which decreases the number of available subgap QP states. All of this leads to a progressive growth of the slope of $dI/dV(V)$ characteristics with decreasing T.

The sudden appearance of excess resistance at $T < T_c$ in overdoped mesas indicates that the superconducting transition here is conventional (i.e., accompanied by an opening of the superconducting gap) and consistent with a single QP tunneling mechanism of interlayer transport.

However, the superconducting transition in underdoped mesas is clearly abnormal. We emphasize again that the appearance of the negative excess resistance and the crossover to T-independent slope reflect the same phenomenon at zero and finite bias, respectively. Therefore, any explanation of the observed unusual T dependence must account for both zero and finite bias behavior.

At a first glance, the lack of excess resistance at $T < T_c$ in underdoped mesas seems consistent with the precursor superconductivity scenario of the pseudogap [3], according to which the gap does not open and QPs do not start to pair at T_c but at much higher T^*. In this case, the lack of excess resistance would simply reflect the lack of dramatic changes in the QP spectrum upon establishment of the global phase coherence at T_c. However, the same argument would make it difficult to explain the abrupt crossover to a T-independent slope. Furthermore, as we noted above, the slope of $dI/dV(V)$ should continue to increase with decreasing T for single QP tunneling. Therefore, the "quantum state" at $T < T_c$ can hardly be explained in terms of single QP tunneling [19].

This conclusion brings us to one possible interpretation of the observed phenomena. If the c-axis transport in underdoped mesas is not solely due to single QPs, it must also involve pairs. The corresponding multiparticle tunneling [23,24] and the multiple Andreev reflection [25,26] processes are well studied for conventional low-T_c junctions. Both processes are similar [26] and are almost T-independent, because they do not rely on the presence of thermally excited QPs but directly involve Cooper pairs from the Fermi level. The multiparticle current occurs via elastic conversion (dissociation or recombination) of Cooper pairs into QPs, while the multiple Andreev reflection process is due to transmutation of a quasielectron into a quasihole with the creation of a Cooper pair and can be universally described in terms of inelastic tunneling in the presence of a time-dependent phase difference due to the ac-Josephson effect [26].

Why would multiparticle processes become more pronounced in underdoped crystals? It is known that the multiparticle current decreases much faster than the single QP current with decreasing the interface transparency [23,26]. Therefore, the single QP dominates over the multiparticle current, unless there are microshorts in the tunnel barrier [24]. The required microshorts in underdoped Bi-2212 could arise from doping-dependent nanoscale inhomogeneity, observed at the surface of Bi-2212 [27,28], which in this case should persist also in the bulk.

Importantly, the multiparticle processes result in an almost T-independent and V-exponential subgap current [23,24], which coincides with our observations; see Fig. 2. Furthermore, the abruptness of the crossover at T_c points towards the coherent multiple Andreev reflection mechanism of the interlayer current, because it requires the time-periodic ac-Josephson effect [26] and, consequently, abruptly disappears simultaneously with the phase coherence at T_c.

Finally, we want to discuss the significance of reported simple TA behavior in the normal state. Although similar behavior can be obtained within the pairing gap scenario, with a fortunate combination of many fitting parameters (but not in the "quantum state" $T < T_c$), the amazing success of the trivial TA description [Eq. (1)], without fitting parameters, deserves further consideration. Such a behavior can be ascribed to the band-structure effect, inelastic tunneling via an impurity [28,29] or elastic tunneling via a resonant [30] state in the tunnel barrier, or Coulomb blocking of tunneling [7]. Importantly, all of them do not involve any gap, nor angular dependence in the QP spectrum, but assume instead that there is some constant blocking barrier for interlayer hopping [18]. This may indicate that the "large" c-axis pseudogap is not a pairing gap and would naturally explain its indifference to such classical depairing factors as T and magnetic field [7].

To conclude, we observed a remarkable crossover from thermal activation to quantum tunnelinglike behavior at $T = T_c$. Surprisingly, the crossover becomes more abrupt in underdoped crystals, characterized by smooth and featureless transition through T_c. As we argued, our data indicate that not only the electronic structure but also the c-axis *transport mechanism* changes with doping: from coherent and directional *single* quasiparticle tunneling in overdoped to a progressively increasing *pair* contribution in underdoped Bi-2212. The latter is apparent only in the phase coherent state at $T < T_c$, is almost T-independent, and is consistent with multiple Andreev reflection mechanism of the interlayer transport.

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*Vladimir.Krasnov@physto.se

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