

## Giant dissipation peak and current effect of in-plane resistance in $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+y}$ single crystals under magnetic fields

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(Received 31 January 2000)

By measuring the in-plane resistance on overdoped  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+y}$  single crystals, we observed a giant dissipation peak below the zero-field superconducting transition temperature when applying an external magnetic field. The temperature dependence of the fields corresponding to zero resistance exhibits an upward curvature which is related to the so-called “anomalous  $H_{c2}$ ” behavior. Furthermore, an obvious current-effect was observed under the dissipation maximum. Intensive data analysis reveals that these phenomena indicate strong evidence for the existence of superconducting islands in the Cu–O planes, which is caused by phase separation and lead to the Josephson-coupling-type dissipative mechanism of in-plane transport in overdoped high temperature superconductors.

### I. INTRODUCTION

Recently, a number of experimental results have extended our understanding of the substantial inhomogeneity in high temperature superconductors (HTS), which seems to be induced by phase separation. In fact, because of the short coherent length in HTS, the intrinsic “granular” feature of the sintered polycrystal has attracted a lot of attention since the discovery of the HTS.<sup>1</sup> For instance, Gerber *et al.*<sup>2</sup> have intensively studied the transport properties of real granular  $LM\text{CuO}$  ( $L=\text{Pr, Nd, Sm, Eu}$ ;  $M=\text{Ce, Th}$ ) system, and found a double-peak superconducting transition under certain magnetic fields. To some extent, this dissipation peak is analogous to the out-of-plane resistance–temperature (RT) curves.<sup>3–5</sup> Moreover, they also suggested that this behavior arose from a combination of quasiparticle and Josephson tunneling between isolated superconducting islands.

One of the possible results induced by the granular feature of superconductors is the anomalous  $H_{c2}(T)$  behavior,<sup>6–8</sup> that is, the broad region of upwards curvature and steep slope as temperature approaching zero, which have been observed in overdoped  $\text{Tl}_2\text{Ba}_2\text{CuO}_y$ ,<sup>9</sup>  $\text{Bi}_2\text{Sr}_2\text{CuO}_y$ ,<sup>10</sup> and  $\text{YBa}_2(\text{Cu}_{0.97}\text{Zn}_{0.03})_3\text{O}_{7-\delta}$ .<sup>11</sup> While in the conventional theory,<sup>12</sup>  $H_{c2}$  is proportional to  $(T_c - T)$  near  $T_c$ , and exhibits downwards curvature. For a long time, this anomaly has been controversial: Some researchers regarded it as an intrinsic feature of HTS (for example, see references in Ref. 13), but since most of the anomalous data was obtained from resistance measurements, it has been questioned that this phenomenon may just come of the inhomogeneity of HTS.<sup>2,7,8</sup> In our previous article,<sup>14</sup> we have discussed the overdoped  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+y}$  ( $x=0.20, 0.25$ ) single crystals by magnetic measurements, and provided a strong evidence of a Josephson-coupling origin for the upward curvature of the so-called  $H_{c2}$ ; furthermore, we argued at the end of the article that phase separation may take place in the overdoped samples and result in isolated superconducting islands embedded in the background of normal metals. In addition, this suggestion was also proposed later in Ref. 7.

In this article, we report a giant dissipation peak of the in-plane resistance on overdoped  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+y}$  single

crystals in magnetic fields (as shown in Fig. 1), a double-peak superconducting transition is also observed under certain fields, as discussed in granular superconductors.<sup>2</sup> Though the general shape of the RT curves is similar to that of the out-of-plane Josephson tunneling junctions (JJ) (Refs. 3–5) and granular system<sup>2</sup>, which implies a Josephson-coupling-type transport mechanism; the results of current effect measurements are different from those in the JJ system, but analogous to the granular system. Therefore, the Josephson coupling may take place between some kind of superconducting clusters, like that in the granular superconductors but not JJ. Further theoretical analysis confirmed this conclusion by fitting the experimental data. Because our experiments were performed on high quality single crystals, we discussed that this is clear evidence of phase separation in overdoped samples, which lead to the appearance of the superconducting islands in Cu–O planes. Another important conclusion is that the so-called  $H_{c2}$  anomaly revealed by resistance measurements may be due to the intrinsic inhomogeneity induced by phase separation in high  $T_c$  superconductors.

Before presenting our data, it may be worthy to take a review of the reports on the dissipation of out-of-plane Josephson tunneling junctions (JJ). Normally, high temperature superconductors can be modeled as stacks of JJ with superconducting  $\text{CuO}_2$  layers embedded in insulating, semiconducting, or semimetallic charge reservoirs. As the most anisotropic compounds,  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}$  single crystals have been intensively studied by the  $c$ -axis transport measurements as a typical stack of JJ.<sup>3–5</sup> Experimental results on these crystals are consistent with each other and show a large peak in the out-of-plane RT curves; moreover, with the increase of field, this peak increased in magnitude and the zero-resistance temperature shifted to lower values.<sup>3–5</sup> In addition, as a feature of JJ, a strong current effect was observed below the zero-field  $T_c$ .<sup>5</sup> Though the mechanism of this phenomenon is still an open question, all the explanations are based on the point by modeling this conduction as a series stack of JJ (see for example, Ref. 3,15). In other words, it has been established that the giant RT peak and current effect in magnetic field are characteristics of the JJ-type dissipation.

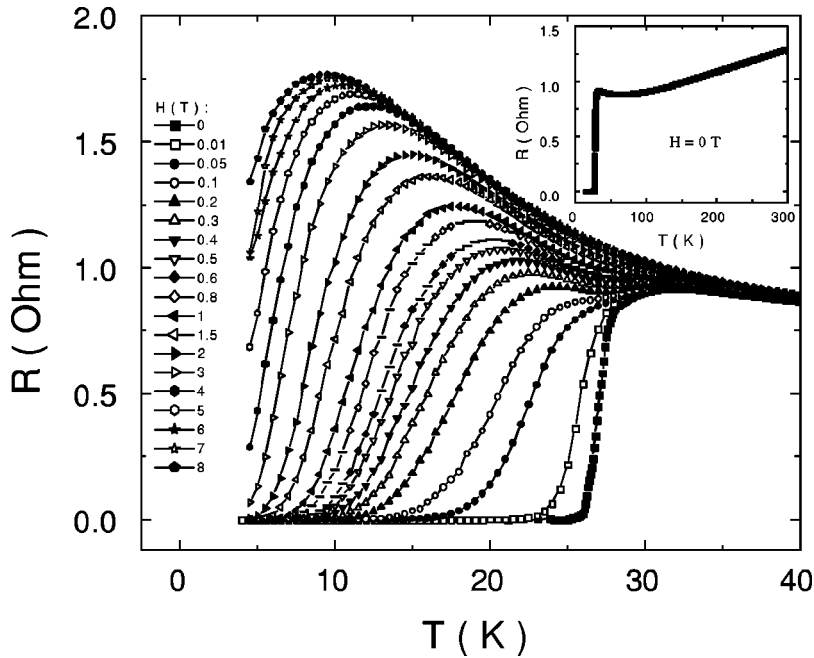


FIG. 1. Temperature dependence of the in-plane resistance on a  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+y}$  ( $x=0.25$ ) single crystal in different magnetic fields. Inset shows the zero-field superconducting transition with the temperature from 14 to 295 K, which is a normal in-plane RT curve.

## II. EXPERIMENT

La-doped  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+y}$  (Bi-2201) single crystals have been grown by the conventional self-flux method using CuO as flux. X-ray diffraction (XRD) patterns and the electron diffraction patterns based on the transmission electron microscopy (TEM) confirmed the excellent crystallinity of our samples. It is also found that the actual La content as determined by energy dispersive x-ray (EDX) analysis depends monotonously on the nominal doping level. Details on sample preparation and characterization have been described in Ref. 16. For the sake of simplicity, in this article we mention only the actual composition. Figure 2 shows the XRD patterns for a crystal with  $x = 0.25$  which is studied below, it can be seen that there are no peaks from any possible impurity, even the diffraction intensity is plotted logarithmically vs the  $2\theta$  angles [as shown in Fig. 2(b)]; besides, the TEM diffraction patterns measured at room temperature are similar to that of the standard Bi-2201 crystals (see for example, Refs. 16,17) without special features.

In this article, we present the data extracted from the transport measurements on crystals with  $x = 0.25$ . It is believed that to substitute  $\text{La}^{3+}$  for  $\text{Sr}^{2+}$  in the system  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+y}$  will reduce the hole numbers in the  $\text{CuO}_2$  planes and thus push the system to the hole-underdoped regime, and the optima doping level is  $x \approx 0.4$ ; thus samples mentioned in this article are hole-overdoped. This is also demonstrated by the clear improvement of both the superconducting fraction and the transition temperature after annealed in flowing Ar gas at  $680^\circ\text{C}$  for 15 h. In addition, we have performed the thermoelectric power (TEP) measurements on the crystal, and the TEP at 290 K is about  $1.38 \mu\text{V/K}$ ; then by using the universal relationship between the TEP at 290 K and the value of hole concentration  $p$  (Ref. 18) ( $p$  is the fraction of holes per Cu atom in the  $\text{CuO}_2$  sheet), we can deduce that  $p$  is about 0.17 when  $x = 0.25$ ,<sup>19</sup> which is in overdoped regime.

Typically, the crystals were cut into bars with rectangular surface dimensions of about  $0.5 \times 2 \text{ mm}^2$  and thickness of

about  $20 \mu\text{m}$ . Silver epoxy was painted on the surfaces to make four electrodes along the bars, and annealed at  $250^\circ\text{C}$  for 15 h; this procedure reduced the contact resistance to about  $1\Omega$ , and ruled out the possibility of any effects from the electrode Joule-heating or insulating contact resistance. The in-plane resistance was measured by a standard four-probe dc technique using the Keithley 220 and 182, and the data was checked to be independent of exact contact configuration by measuring several crystals. Magnetic fields were oriented in the  $c$ -direction and varied from 0 to 8 T supplied

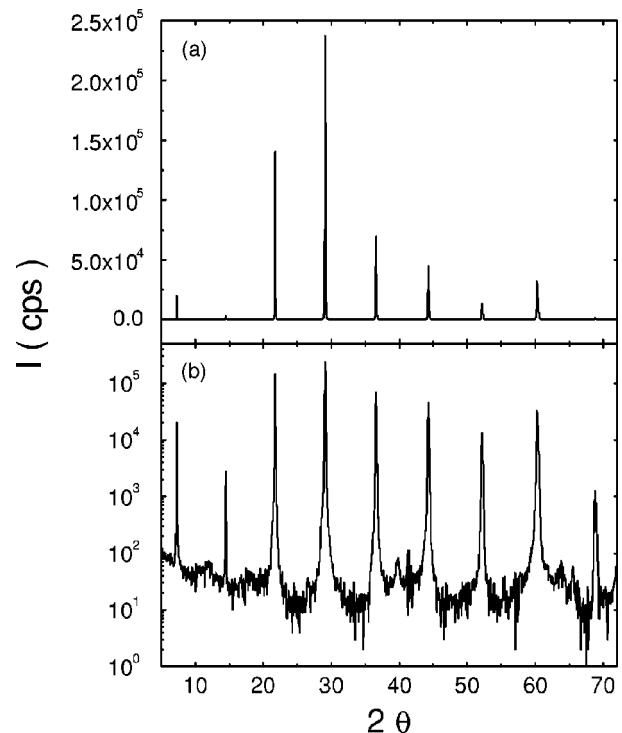


FIG. 2. (a) XRD patterns of the  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+y}$  ( $x=0.25$ ) crystal. (b) The same XRD patterns with diffraction intensity plotted logarithmically vs the  $2\theta$  angles.

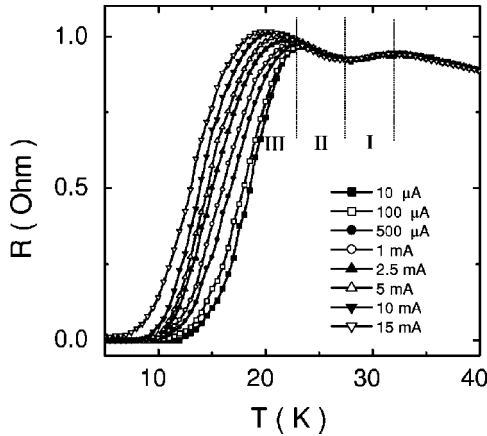


FIG. 3. Temperature dependence of the in-plane resistance measured on the same crystal under external magnetic field of 0.3 T, and the current level from 10  $\mu\text{A}$  to 15 mA. The current effect can also be seen in other applied fields as shown in Fig. 4.

by an Oxyford commercial superconducting magnet.

Figure 1 shows the temperature dependence of the in-plane resistance in different magnetic fields; resistance in normal states is almost the same for each field. In order to take an overview, we present in the inset of Fig. 1 the superconducting transitions with the temperature from 14 to 295 K without external field, which is a normal in-plane zero-field transition. Obviously, the primary feature of Fig. 1 is the giant dissipation peak under fields, the resistance of the dissipation maxima has even exceeded that at 290 K (inset of Fig. 1) when the field is as low as 1.5 T. Moreover, double peak in RT curves, as mentioned by Gerber *et al.*,<sup>2</sup> is observable under the fields below 0.6 T, approximately.

At first sight, the RT curves shown in Fig. 1 are similar to those of the JJ system,<sup>3-5</sup> implying the existence of some kind of Josephson coupling. In order to make this point clear, we studied the current effect on the superconducting transitions. As a representation, some of the RT curves, measured on the same crystal at 0.3 T field by using different currents, are presented in Fig. 3. The first (high-temperature) resistance peak was found to be unchanged by the current increase from 10  $\mu\text{A}$  to 15 mA, in contrast to the low-temperature one which shifted to lower temperatures clearly. The excellent reproducibility of the first transition demonstrates the comparability of these curves, and we did not show the data for current exceeding 15 mA because too large current will result in a great heating effect, leading to irrelevant data. Moreover, the same phenomenon can be seen under other magnetic fields (as shown in Fig. 4), and for the single-peak curves measured in high fields, current effect is available just below the dissipation maxima. Therefore, the giant peaks in the single-peak curves under high fields and the low-temperature peaks in the double-peak curves under low fields correspond to the beginning points of current effect, implying the beginning of Josephson coupling.

### III. DISCUSSION

In spite of the analogous shape of the RT curves, the current effect shown here, which took place only below the low-temperature transition, is different from that of the

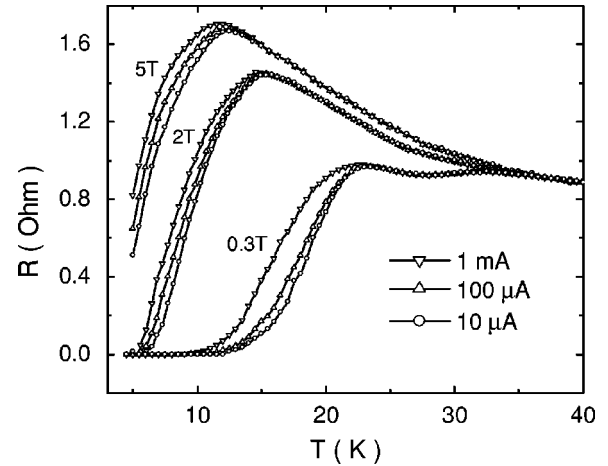


FIG. 4. The in-plane resistance-temperature curves under external magnetic field of 0.3 T, 2 T, and 5 T. Three different current level are applied, and the current effect takes place just below the dissipation maxima under high fields and the second maxima under low fields, implying the beginning points of Josephson coupling.

*c*-axis JJ system, which shows an obvious current effect just below the zero field  $T_c$  (see for example, Ref. 5); while it is qualitatively similar to what have been observed in granular superconductors.<sup>2</sup> This is understandable because the measurements performed in this work were in-plane but not out-of-plane transport; Josephson coupling in this case may take place between some kind of superconducting clusters, like that in the granular superconductors, but not between Cu-O layers, like that in JJ systems. Otherwise, grains in granular superconductors must be much larger<sup>2</sup> than the superconducting clusters in our crystals, thus leading to the quantitative differences of current effect in these two systems, that is, current effect and the magnitude of the first drop are much weaker in our crystals compared with samples built of large grains.<sup>2</sup> Therefore, we explain the double-peak curves by regarding the first drop (part 1 in Fig. 3) as the appearance of superconducting clusters and the second drop (part 3 in Fig. 3) as the beginning of Josephson coupling between these clusters. Thus the intracluster superconducting transition and intercluster Josephson coupling lead to the two drops in the resistance-temperature curves. By applying high fields, the first peak is smeared out bit-by-bit, and resistance increases dramatically before the steep drop to zero (part 2 in Fig. 3), which may be governed by the single electron tunneling between superconducting clusters as suggested in Ref. 2. Besides, no matter what the magnetic field is, the resistance will drop to zero when all the superconducting clusters are coupled together; that is, the zero-resistance point should correspond to the critical field at which the bulk superconducting state is established.

This explanation has been confirmed by fitting the data with the phenomenological theory proposed by Geshkenbein, Ioffe, and Millis<sup>8</sup> (shown in Fig. 5). Originally, this theory explains the upward curvature of  $H_{c2}(T)$  appearing in overdoped Ti-2201 system as the result of a bulk superconducting phase coherence formed through Josephson coupling between some superconducting clusters, perhaps caused by inhomogeneous oxygen concentration. According to this theory, the critical field, under which a bulk phase coherence is established, is described by

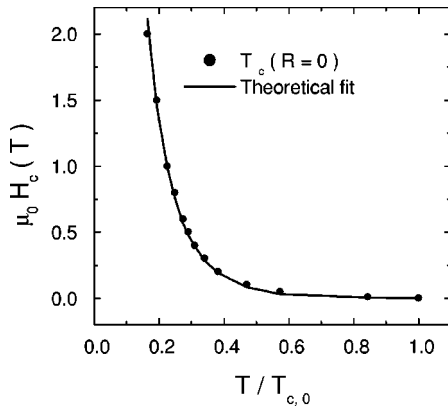


FIG. 5. Fields corresponding to the zero-resistance points in RT curves. The line represents a theoretical fit with the Eq. (1) (see text).

$$H_c = H_0 \frac{T_c}{T} \exp\left(-\frac{T}{T_0}\right), \quad (1)$$

where  $T_0 = v_F/2\pi d$ ,  $v_F$  is the Fermi velocity,  $d$  is the average distance between the clusters, and  $H_0$  is a parameter related to the configuration of the clusters and  $v_F$ . As mentioned above, in the RT curves, superconducting clusters are coupled together into a bulk superconductor at the point of zero resistance. Therefore, in Fig. 5, we show the temperature dependence of the critical field, corresponding to the zero resistance on each of the curves in Fig. 1, and the solid line in Fig. 5 represents a fit to Eq. (1) with  $T_0 = 3.5$  K and  $H_0 T_c = 2.8 \times 10^5$  TK. Obviously, the theoretical curve gives a remarkably good description of the experimental data. The excellent fit of the theoretical curve to the experimental data demonstrates the Josephson coupling origin for the upward curvature of the so-called upper critical fields at zero-resistance point in transport measurements. This result is consistent with what has been concluded from magnetic measurements,<sup>14</sup> and gives a more direct evidence that the  $H_{c2}$  anomaly determined by resistive measurement in some overdoped samples may not be an intrinsic property of HTS, but a reflection of the substantial inhomogeneity in HTS. However,  $T_0$  deduced from magnetic measurements is 1.85 K,<sup>14</sup> different from the 3.5 K resulting from RT curves here. This may be due to two possible reasons: (i) the detecting methods are different: in Ref. 14,  $T(H_c)$  is defined as the onset point of the second transition in magnetization-temperature curves, while in this article, it is obtained from the zero-resistance point; (ii) the characteristics of the crystals are different: it has been widely accepted that the nonstoichiometry of Bi and Sr atoms, which is common in Bi-2201 system, can lead to distinct differences on physical properties, even though the La contents are fixed.<sup>16,20</sup> Our crystals, which are grown by the self-flux method, cannot be free from this nonstoichiometry; nevertheless, this did not affect the qualitative results.

It may be worthy to note that while the critical fields correspond to the dissipation maxima in RT curves, the  $T_c$  onset also exhibits a positive curvature with the decrease of temperature. Furthermore, we measured the RT curves under magnetic fields on  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+y}$  single crystals and got the same results on some crystals. In fact, the in-plane dissi-

pation maxima in Bi-2212 system has been reported by Hsu *et al.*,<sup>21</sup> but we have not seen the reported annealing effect very clearly, and their explanation based on the vortex-line distortions can not interpret the current effect shown in this article.

Another question is how the superconducting clusters are formed. The eventual answer to this question is of course beyond the context of this article; however, because these experiments were performed on high quality single crystals, we can imagine that some kind of phase separation led to the appearance of the superconducting clusters. In another article,<sup>22</sup> by studying the differences on the time evolution of the two coexistent transitions, we have concluded that this phase separation cannot be attributed to any chemical reason or vortex motion, but to some intrinsic driving force, such as the electronic-driven phase separation. Therefore, what we have reported here may give further evidence of the intrinsic inhomogeneity induced by the electronic phase separation in overdoped high-temperature superconductors.

#### IV. CONCLUSION

In summary, we have measured the in-plane resistance of overdoped  $\text{Bi}_2\text{Sr}_{2-x}\text{La}_x\text{CuO}_{6+y}$  single crystals. A giant dissipation peak is observed under magnetic field, which seems, at first sight, like the transport property of out-of-plane Josephson tunnel junctions,<sup>3-5</sup> indicating a Josephson-tunneling dissipation mechanism; in addition, double-peak transitions are observable under certain fields as reported in granular superconductors.<sup>2</sup> While experimental results on current effect show that the Josephson coupling occurring in our samples is different from that due to the  $c$ -axis coherence,<sup>5</sup> but analogous to that in the granular superconductors,<sup>2</sup> suggests that the coupling must take place between the superconducting islands in the Cu-O plane. Therefore, consistent with the magnetic measurements published before,<sup>14</sup> we explain the transport properties as due to the presence of intrinsic inhomogeneity in our overdoped crystals with superconducting clusters embedded in normal metal, which may be caused by electronic phase separation.<sup>22</sup> In this scenario, the two drops in double-peak RT curves correspond to intracluster superconducting transition and intercluster Josephson coupling, respectively, and the current effect between the maximum and zero-resistance indicates that the superconducting clusters are coupled together into a bulk superconductor.

This explanation has been confirmed by fitting the experimental data with a phenomenological theory based on Josephson coupling between small grains with  $T_c$  higher than the bulk.<sup>8</sup> Another important conclusion is also easily deduced from this data fitting; that is, the so-called ‘‘ $H_{c2}$  anomaly’’ discovered in resistance measurements may be only a reflection of the substantial inhomogeneity in high-temperature superconductors.<sup>7,14,8</sup>

#### ACKNOWLEDGMENTS

We are very grateful to Professor Lu Li for his help on transport measurements, and Y. M. Ni for technical assistance. This work was supported by NSFC within the project 19825111.

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