Insulator-to-Metal Crossover in the Normal State of $La_{2-x}Sr_xCuO_4$ Near Optimum Doping

G. S. Boebinger, Yoichi Ando,* and A. Passner
Bell Laboratories, Lucent Technologies, 700 Mountain Avenue, Murray Hill, New Jersey 07974

T. Kimura, M. Okuya, J. Shimoyama, and K. Kishio
Department of Applied Chemistry, University of Tokyo, Hongo, Bunkyo-ku, Tokyo 113, Japan

K. Tamasaku, N. Ichikawa, and S. Uchida
Superconductivity Research Course, University of Tokyo, Yayoi, Bunkyo-ku, Tokyo 113, Japan

(Received 29 July 1996)

A 61-T pulsed magnetic field suppresses superconductivity in $La_{2-x}Sr_xCuO_4$ single crystals and reveals an insulator-to-metal (IM) crossover for both in-plane resistivity ρ_{ab} and c-axis resistivity ρ_c at a Sr concentration near optimum doping ($x \simeq 0.16$). The IM transition is unusual in that *all* underdoped samples ($x < 0.16$) show low-temperature insulating behavior, even in samples with linear-T ρ_{ab} above T_c and apparently large $k_F l$. [S0031-9007(96)01773-5]

PACS numbers: 74.25.Dw, 74.25.Fy, 74.72.Dn

The anisotropic normal-state resistivity of the high-temperature superconducting cuprates exhibits a peculiar evolution as the carrier concentration is increased through the superconducting phase [1,2]. As the carrier concentration is increased from the undoped insulator, by Sr doping in the case of $La_{2-x}Sr_xCuO_4$ (LSCO), the insulating phase ("I" in the inset of Fig. 1) gives way to superconductivity (SC). If the cuprate is sufficiently overdoped that superconductivity is destroyed, both the in-plane resistivity ρ_{ab} and c-axis resistivity ρ_c exhibit "metallic" ($d\rho/dT > 0$) behavior down to the lowest experimental temperatures and share some of the properties of an anisotropic three-dimensional Fermi liquid [1–5]. However, the low-temperature limiting behavior of the crossover from insulator-to-metal (IM) is hidden by the existence of the superconducting phase.

Our goal is to study this IM crossover in the low-temperature normal-state resistivity as a function of carrier concentration in the absence of superconductivity. Existing data on LSCO above T_c suggest that increasing carrier concentration drives ρ_{ab} metallic before ρ_c [2,3]. In general, near optimum doping, ρ_{ab} in the cuprates is metallic and displays the well-known linear-T temperature dependence, while ρ_c exhibits "insulating" ($d\rho/dT < 0$) behavior [1]. This contrasting behavior between ρ_{ab} and ρ_c has been cited as evidence of an unusual non-Fermi-liquid ground state in the high-T_c cuprates [6]. Whether or not this contrasting behavior generally extends to low temperatures is an open issue.

A standard technique to extend normal-state measurements to lower temperatures is to suppress superconductivity by chemical substitution [7–9]. The resulting increased disorder tends to increase the normal-state resistivity and offers an opportunity to study localization [1]. In an ordinary metal, as disorder is increased, insulating behavior is expected once the mean free path ℓ between scattering events becomes shorter than the wavelength of an electron at the Fermi energy, i.e., once $k_F\ell < 1$, where k_F is the Fermi wave vector [10]. In a free electron model for a layered two-dimensional (2D) material, the magnitude of ρ_{ab} directly gives $k_F\ell = hc_o/\rho_{ab}e^2$, where c_o is the interlayer distance (6.5 Å in LSCO).

In the underdoped high-T_c cuprates, chemical substitution (for example, doping Zn onto the Cu site) suppresses superconductivity and induces a crossover from superconducting to insulating behavior. The observed crossover is similar to that of conventional 2D superconductors: it occurs at ρ_{ab} corresponding to $\sim h/4e^2$ per CuO layer ($k_F\ell \sim 4$) [1,8,9]. This suggests that Cooper pair localization [11] underlies the disorder-induced insulating behavior in underdoped high-T_c cuprates.

Because we want to study the low-temperature evolution of ρ_{ab} and ρ_c in samples without deliberate chemical substitution to suppress superconductivity, we utilize a 61–T pulsed magnet [12]. We study LSCO because single crystal samples are available over a wide range of doping and because 61-T magnetic fields can suppress superconductivity to very low temperatures in LSCO even near optimum doping. The inset of Fig. 1 shows the measured T_c (midpoint) versus nominal Sr concentration of the single crystals studied, with the superconducting transition width indicated by the vertical bars. The samples with Sr concentrations $x = 0.08$, 0.13, 0.17, 0.18, and 0.22 were grown by the Kishio group [13], while those with $x = 0.12$ and 0.15 were grown by the Uchida group [2]. The crystallographic axes are determined and the samples are cut into platelets (typically $2.5 \times 1.5 \times 0.5$ mm^3) for standard four-terminal resistivity measurements of either ρ_{ab} or ρ_c. The temperature dependence of the zero-field resistivity is measured using a lock-in amplifier driven at 17 Hz (solid lines in Figs. 1 and 2).

For each magnetic-field pulse, the sample temperature is stabilized at a desired temperature within the experimental range of 0.65 to 200 K. The current density

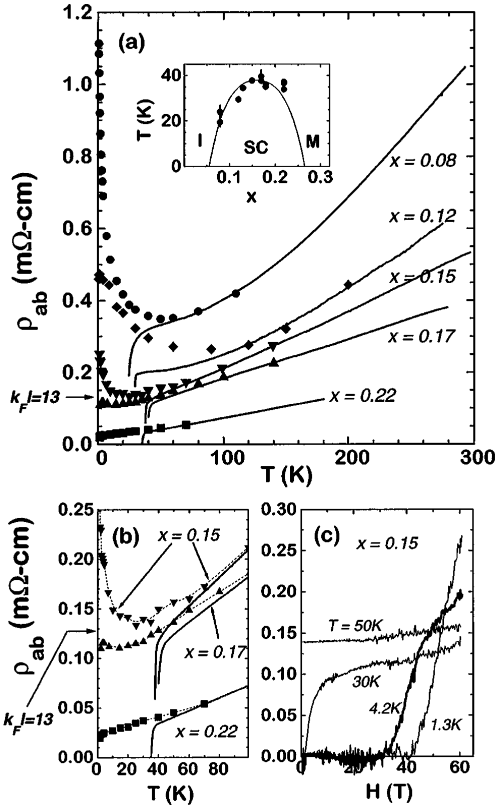

FIG. 1. Inset gives the T_c (midpoint) versus Sr concentration, x, for the $La_{2-x}Sr_xCuO_4$ single crystals studied. The size of the circles gives the typical superconducting transition width (90%–10%), where vertical bars indicate broader transitions. (a) ρ_{ab} for five samples of different x. The lines/symbols are 0 T/60 T data for $x = 0.08$ (circles), 0.12 (diamonds), 0.15 (down triangles), 0.17 (up triangles), and 0.22 (squares); (b) close-up of the insulator-to-metal crossover at $x \simeq 0.16$ and apparent $k_F l \sim 13$; (c) raw ρ_{ab} versus H traces for the $x = 0.15$ sample, showing clear insulating behavior at $H = 60$ T for $T < 30$ K.

(typically 1.5 A/cm^2) is kept within the ohmic range of the normal-state resistivities. The magnetic field H is applied along the c-axis to most effectively suppress superconductivity, and the isothermal $\rho(H)$ is recorded by a transient digitizer monitoring the fast (10 μs) output of a lock-in amplifier driven at $\sim$120 kHz. Despite the transient nature of the magnetic field, the samples are sufficiently small that the data are not adversely affected by eddy-current heating during the relatively long $\sim$100 ms magnetic-field pulse.

To determine the temperature dependence of the normal-state resistivity below T_c, each sample is subject to a series of (typically twenty) 61-T magnetic-field pulses at different temperatures. The symbols in Figs. 1 and 2 represent the ρ_{ab} and ρ_c measured at 60 T. For most of the samples studied, the magnetoresistance of the normal state is small on the scale of Figs. 1 and 2, as evidenced by the general accord between the zero-field and 60-T data well above T_c. The scatter in the 60-T data [particularly evident in Fig. 1(b)] results from the (few μV) noise in the pulsed-magnetic-field environment. Despite these uncertainties, the raw $\rho_{ab}(H)$ data for $x = 0.15$ [Fig. 1(c)] clearly show insulating behavior in the normal state for $T < 30$ K, whereas the $x = 0.17$ data appear to saturate at a finite conductivity. For many samples near optimum doping ($x \simeq 0.16$), even 61–T is unable to completely suppress superconductivity and the onset of the superconducting transition is visible at the lowest temperatures in Figs. 1 and 2.

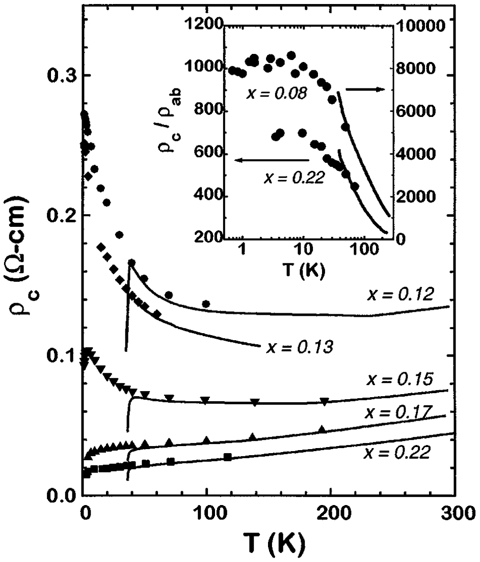

FIG. 2. ρ_c for five samples of different x. The lines/symbols are 0 T/60 T data for $x = 0.12$ (circles), 0.13 (diamonds), 0.15 (down triangles), 0.17 (up triangles), and 0.22 (squares). The $x = 0.13$ data are from Ref. [16]. The inset shows the low-temperature saturation of the anisotropy ratio.

There are several striking features in the data of Fig. 1. The first is that all samples exhibit metallic behavior above T_c in zero magnetic field and yet *every one* of the underdoped samples exhibits insulating behavior once superconductivity is suppressed. Second, the IM crossover in ρ_{ab} [expanded view in Fig. 1(b)] occurs between $x = 0.15$ and 0.17, very near the Sr concentration corresponding to optimum T_c. In every underdoped sample, except at $x = 0.12$ [14], both ρ_{ab} and ρ_c show no evidence of saturation at low temperatures and diverge as the logarithm of the temperature [16]. This suggests an unusual insulating ground state in LSCO *which extends up to optimum doping.*

Figure 2 contains $\rho_c(T)$ data which show that the IM crossover in ρ_c occurs between $x = 0.15$ and 0.17, just as in ρ_{ab}. In every LSCO sample studied, the low temperature ρ_{ab} and ρ_c are either both insulating or both metallic, despite the contrasting behavior which is often observed above T_c. There is further evidence that the low-temperature ρ_{ab} and ρ_c are related; the anisotropy ratio ρ_c/ρ_{ab} saturates at low temperatures for *all* Sr concentrations, independent of whether the normal-state resistivities exhibit insulating or metallic behavior. This is represented by the data in the inset of Fig. 2 for $x = 0.08$ and 0.22, which are consistent with previous data [16] on underdoped, insulating LSCO.

The discussion has focused on the evolution of the normal-state resistivity with increasing carrier concentration: ρ_{ab}, ρ_c, and the anisotropy ratio all decrease, while ρ_{ab} and ρ_c both undergo an IM crossover. Ordinarily, increasing Sr doping should increase not only carrier concentration but also the disorder in the samples. Experimentally, the rapidly decreasing resistivity with Sr doping suggests, however, that the effects of increasing the carrier concentration dominate. It is interesting to note that insulating behavior is observed in samples with very low ρ_{ab}, corresponding to $k_F\ell$ apparently as large as 13, using the free electron model discussed previously (arrow in Fig. 1). Such large values of $k_F\ell$ would ordinarily accompany metallic behavior. It might be the case that the intense magnetic field is inducing the insulating behavior (the magnetic length is only 33 Å at 60 T); however, the small normal state magnetoresistance suggests rather that the magnetic field is "unveiling" the intrinsic normal state behavior [16]. Insulating behavior in samples with $k_F\ell$ as large as 13 would suggest that the effects of disorder are enhanced in LSCO, consistent with a general prediction for non-Fermi liquids that arbitrarily small disorder will lead to insulating behavior in both ρ_{ab} and ρ_c [17]. One could argue that our estimation of $k_F\ell$ is incorrect; however, the free electron 2D formula is the most obvious simple model available for estimating $k_F\ell$, given the large magnitude of the anisotropy ratio in LSCO.

Figure 3 summarizes the low-temperature behavior of all samples studied as a function of Sr concentration.

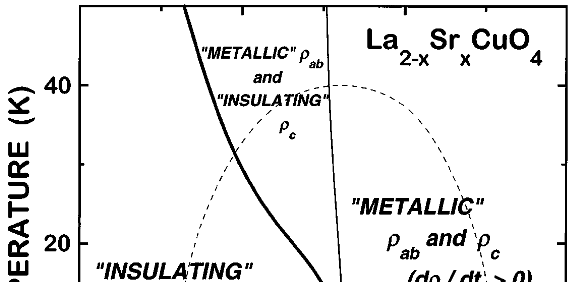

FIG. 3. Schematic diagram of the behavior of the normal state resistivity in the absence of superconductivity versus Sr concentration. The dashed line gives $T_c(x)$ in zero magnetic field. An insulator-to-metal crossover occurs in *both* ρ_{ab} and ρ_c near optimum doping at low temperatures.

(I) The IM crossover occurs in all three directions, in that both ρ_{ab} and ρ_c exhibit the same low-temperature behavior for all Sr concentrations studied. Thus, at least in LSCO [18], the famous contrasting behavior of linear-T ρ_{ab} and insulating ρ_c does not persist to low temperatures in the absence of superconductivity. The contrasting behavior has been cited as evidence of quasiparticle confinement in a 2D Luttinger liquid [20]. In this picture, the observed insulating $\rho_{ab} \propto \rho_c$ could result from residual disorder in the underdoped samples [21].

(II) The low-temperature insulating behavior in the normal state is unusual, as it occurs only in underdoped samples. Although these samples are superconducting in zero magnetic field, we believe that this insulating behavior is not due to Cooper pair localization [8,9], because Cooper pair localization would not exist above H_{c2} [22]. We note that Refs. [8,9] study the disorder-driven superconductor-to-insulator transition, which is not directly related to the carrier-concentration-driven IM transition which we report here.

Feature (I) is reminiscent of behavior reported for Nd-substituted LSCO [23]. In $\mathrm{La}_{1.6-x}\mathrm{Nd}_{0.4}\mathrm{Sr}_x\mathrm{CuO}_4$, the Nd substitution induces a low-temperature structural phase transition to a tetragonal phase which is similar to that reported near $x = 1/8$ in $\mathrm{La}_{2-x}\mathrm{Ba}_x\mathrm{C}_4$. In this low-temperature phase, both ρ_{ab} and ρ_c are insulating, even in overdoped samples with apparent $k_F\ell$ as large as 24. Near $x = 1/8$ in $\mathrm{La}_{1.6-x}\mathrm{Nd}_{0.4}\mathrm{Sr}_x\mathrm{CuO}_4$, charge and spin ordering has been observed [15]. It is possible that charge ordering could underlie the unusual insulating behavior which we observe in *all* underdoped LSCO samples.

We note also that an as-yet unexplained, logarithmically divergent resistivity has been observed in granular films of NbN, a conventional superconductor [24]. Given the possibility of phase segregation in the LSCO system, there might be a link between the unusual insulating behavior in LSCO and that in granular NbN films; however, this would seem to require phase segregation throughout and only in the underdoped regime of LSCO.

Finally, features (I) and (II) together suggest that the low-temperature normal-state IM crossover in LSCO is linked to the superconducting phase, since it occurs at nearly the same carrier concentration as optimum T_c. This IM crossover in LSCO joins a growing list of crossover behaviors which occur in the normal state near optimum doping: in NMR [25], neutron scattering [26], optical conductivity [2728], specific heat [29], thermoelectric power [30], susceptibility [31], resistivity [31,32], the Hall coefficient [33], and photoemission [34]. Many phenomena which are unique to underdoped samples have been linked to the pseudogap, a suppression in the excitation spectrum of the normal state above T_c, although there is ongoing debate whether single-layer materials (such as LSCO) show a pseudogap at all. Certainly, experimental evidence for the pseudogap is stronger for double-layer materials and theoretical arguments suggest that a gap in the spin excitation spectrum can only be formed in double-layer systems [35]. We also point out that the onset of the pseudogap occurs at temperatures well above T_c and results in a *decrease* in ρ_{ab} [31,32]. Thus, the link, if any, between the pseudogap and the peculiar insulating behavior is not immediately obvious.

In conclusion, we have studied the evolution of the low-temperature ρ_{ab} and ρ_c with carrier concentration in LSCO with the assistance of a 61-T pulsed magnetic field. Both ρ_{ab} and ρ_c cross from insulating to metallic behavior near the Sr concentration $x \simeq 0.16$ corresponding to optimum T_c. Low-temperature insulating behavior is observed in all underdoped samples, even in those exhibiting apparently large $k_F\ell$ and ρ_{ab} which depends linearly on temperature for all $T > T_c$.

---

*Present address: Central Research Institute of Electric Power Industry, Komae, Tokyo 201, Japan.

[1] For reviews, see Y. Iye, in *Physical Properties of High Temperature Superconductors III,* edited by D.M. Ginsberg (World Scientific, Singapore, 1991); S.L. Cooper and K.E. Gray, in *Physical Properties of High Temperature Superconductors IV,* edited by D.M. Ginsberg (World Scientific, Singapore, 1994).

[2] Y. Nakamura and S. Uchida, Phys. Rev. B **47**, 8369(1993), and references therein.

[3] T. Kimura, S. Miyasaka, H. Takagi, K. Tamasaku, H. Eisaki, S. Uchida, K. Kitazawa, M. Hiroi, M. Sera, and N. Kobayashi, Phys. Rev. B **53**, 8733 (1996).

[4] T. Manako, Y. Shimakawa, Y. Kubo, and H. Igarashi, Physica (Amsterdam) **190C**, 62 (1991).

[5] H.M. Duan, W. Kiehl, C. Dong, A.W. Cordes, M.J. Saeed, D.L. Viar, and A.M. Hermann, Phys. Rev. B **43**, 12 925 (1991).

[6] P.W. Anderson, Science **256**, 1526 (1992).

[7] T.R. Chien, Z.Z. Wang, and N.P. Ong, Phys. Rev. Lett. **67**, 2088 (1991).

[8] Y. Fukuzumi, K. Mizuhashi, K. Takenaka, and S. Uchida, Phys. Rev. Lett. **76**, 684 (1996).

[9] D.J.C. Walker, A.P. Mackenzie, and J.R. Cooper, Phys. Rev. B **51**, 15 653 (1995).

[10] N.F. Mott, *Metal-Insulator Transitions,* 2nd ed. (Taylor & Francis, London, 1990), Chap. 1.

[11] M.P.A. Fisher, G. Grinstein, and S.M. Girvin, Phys. Rev. Lett. **64**, 587 (1990).

[12] G.S. Boebinger, A. Passner, and J. Bevk, Physica (Amsterdam) **201B**, 560 (1994).

[13] T. Kimura, K. Kishio, T. Kobayashi, Y. Nakayama, N. Motohira, K. Kitazawa, and K. Yamafuji, Physica (Amsterdam) **192C**, 247 (1992).

[14] $x = 0.12$ is near $x = 1/8$, where charge and spin ordering [15], as well as anomalies in T_c and magnetoresistance [3], are observed. In our $x = 0.12$ samples, ρ_{ab} exhibits a large magnetoresistance extending well above T_c and both ρ_{ab} and ρ_c tend to saturate at the lowest temperatures.

[15] J.M. Tranquada, J.D. Axe, N. Ichikawa, Y. Nakamura, S. Uchida, and B. Nachumi, Phys. Rev. B **54**, 7489 (1996), and references therein.

[16] Y. Ando, G.S. Boebinger, A. Passner, T. Kimura, and K. Kishio, Phys. Rev. Lett. **75**, 4662 (1995).

[17] G. Kotliar and C.M. Varma, Physica (Amsterdam) **167A**, 288 (1990).

[18] Recent measurements on $Bi_2(Sr,La)_2CuO_y$ [19] have found that the contrasting behavior (metallic ρ_{ab} and insulating ρ_c) can persist down to $T = 0.65$ K $(T/T_c \sim 0.05)$.

[19] Y. Ando, G.S. Boebinger, A. Passner, N.L. Wang, C. Geibel, and F. Steglich, Phys. Rev. Lett. **77**, 2065 (1996).

[20] D.G. Clarke, S.P. Strong, and P.W. Anderson, Phys. Rev. Lett. **74**, 4499 (1995), and private communication.

[21] P.W. Anderson, T.V. Ramakrishnan, S. Strong, and D.G. Clarke (to be published).

[22] For a review, see A.F. Hebard, in *Strongly Correlated Electronic Materials,* edited by K.S. Bedell, Z. Wang, D.E. Meltzer, A.V. Balatsky, and E. Abrahams (Addison-Wesley, New York, 1994).

[23] Y. Nakamura and S. Uchida, Phys. Rev. B **46**, 5841 (1992).

[24] R.W. Simon, B.J. Dalrymple, D. Van Vechten, W.W. Fuller, and S.A. Wolf, Phys. Rev. B **36**, 1962 (1987).

[25] For a review, see C. Slichter, in *Strongly Correlated Electronic Materials,* edited by K.S. Bedell, Z. Wang, D.E. Meltzer, A.V. Balatsky, and E. Abrahams (Addison-Wesley, New York, 1994).

[26] J. Rossat-Mignod, L.P. Regnault, C. Vettier, P. Bourges, P. Burlet, J. Bossy, J.Y. Henry, and G. Lapertot, Physica (Amsterdam) **185–189C**, 86 (1991).

[27] L.D. Rotter, Z. Schlesinger, R.T. Collins, F. Holtzberg, C. Field,U.W. Welp, G.W. Crabtree, J.Z. Liu, Y. Fang, K.G. Vandervoort, and S. Fleshler, Phys. Rev. Lett. **67**, 2741 (1991).

[28] C.C. Homes, T. Timusk, R. Liang, D.A. Bonn, and W.N. Hardy, Phys. Rev. Lett. **71**, 1645 (1993).

[29] J.W. Loram, K.A. Mirza, J.R. Cooper, and W.Y. Liang, Phys. Rev. Lett. **71**, 1740 (1993).

[30] J.L. Tallon, J.R. Cooper, P.S.I.P.N. de Silva, G.V.M. Williams, and J.W. Loram, Phys. Rev. Lett. **75**, 4114 (1995).

[31] B. Batlogg, H.Y. Hwang, H. Takagi, R.J. Cava, H.L. Kao,and J. Kwo, Physica (Amsterdam) **235C–240C**, 130 (1994), and referencess therein.

[32] T. Ito, K. Takenaka, and S. Uchida, Phys. Rev. Lett. **70**, 3995 (1993).

[33] H.Y. Hwang, B. Batlogg, H. Takagi, H.L. Kao, J. Kwo, R.J. Cava, and W.F. Peck, Jr., Phys. Rev. Lett. **72**, 2636 (1994).

[34] D.S. Marshall, D.S. Dessau, A.G. Loeser, C-H. Park, A.Y. Matsuura, J.N. Eckstein, I. Bozovic, P. Fournier, A. Kapitulnik, W.E. Spicer, and Z.X. Shen, Phys. Rev. Lett. **76**, 4841 (1996).

[35] B.L. Altshuler, L.B. Ioffe, and A.J. Millis, Phys. Rev. B **53**, 415 (1996), and references therein.