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Systematic evolution of electron localization in $Bi_2Sr_2CuO_y$

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Abstract. The electronic transport properties in $Bi_2Sr_2CuO_y$ crystals have been investigated with the focus on the localized behaviours at low temperature. It is found that in the passage of transition from metallic to insulating states the samples first show two-dimensional weak-localization behaviour, and cross over to the variable-range hopping conduction corresponding to the strong-localization regime. We analyse the experimental results and suggest that the metal–insulator transition is caused by the effects of both disorder in the lattice and electron correlations.

The metal–insulator (M–I) transition is an important topic in condensed matter physics [1]. Apart from the M–I transition associated with the structure instability in the one-dimensional electron system, there are two types of well established M–I transition. One is the Anderson transition, which results from the disordering of the potentials. Electrons become localized in potential fluctuations if the randomness exceeds a certain limit. The other one is the Mott–Hubbard transition, which is caused by the electron correlations. Because of the large on-site electron repulsion, it is energetically unfavourable for electrons to hop from one site to another and create doubly occupied sites. A Mott–Hubbard gap is caused in the Mott insulator. The gap is assumed to vanish at the transition. Due to the discovery of high-T_c superconducting cuprates, there has been increased interest in exploring further the passage from the insulator to the metallic state. A noticeable characteristic of high-T_c cuprates is that the superconducting phase adjoins the antiferromagnetic insulating phase and appears in regions having a rather strong electronic correlation. The normal state physical properties differ considerably from those of usual metals. Therefore, the discovery of high-T_c cuprates provides a good opportunity to study the M–I transition in highly correlated electronic system. Besides, understanding the evolution of the electronic properties in the process of the transition from metal to insulator is also helpful to the understanding of high-T_c superconductivity. In the past several years, although there have been considerable experimental and theoretical explorations of the nature of this M–I transition, our understanding is far from complete. Experimentally, the electrical properties in the insulating and metallic phases have been extensively studied, but comparatively less is known yet about the critical regime of the M–I transition. For many cuprate systems, even in the process of the M–I transition the charge transport behaviours at low temperature were cut off and not observed due to the occurrence of superconductivity. In the insulating regime a hopping conduction was generally observed, which naturally led one to conclude that the transition was of Anderson type [2–6]. However, it seems that the effect of electronic

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correlation is seldom stressed in the transition. In the present work we present a systematic experimental exploration of the electronic transport in selected $Bi_2Sr_2CuO_y$ crystals which cover the transition from metallic to insulating conduction. The advantage in studying this Bi-2201 system is that the superconducting transition temperature is much lower compared with other high-T_c systems. In the case of superconductivity being supressed by reduction of carrier concentration, the electronic transport behaviour in the low-temperature regime can be observed clearly. Our experiments show that, in the passage from the metallic to the insulating state, the crystals first exhibit two-dimensional weak-localization behaviour at low temperature, and cross over to the variable-range hopping (VRH) conduction corresponding to the strong-localization regime. In spite of the transition following a form of Anderson type, our analysis strongly suggests that the electron correlation plays an important role in the M–I transition. We suggest that both electronic correlation and the disorder are present, and enhance each other's effect. Some insight about the M–I transition in correlation electron system is presented.

The $Bi_2Sr_2CuO_y$ crystals were grown from a copper-oxide-rich melt in Al_2O_3 crucibles. Typical dimensions are 3 mm × 2 mm × (10–40μm). The resistances of the crystals with or without the magnetic field were measured from the standard DC four-probe method. To ensure low contact resistance, silver electrodes were evaporated before the copper leads were softly soldered. A current of 100 μA from the Keithley 220 current source, with reversal controlled by computer, was passed through the sample, and the voltage was measured with a Keithley 181 nanovoltmeter. The magnetic field was produced by a superconducting

For different crystals in the experiment, even if they grew from the same crucible, their conduction behaviours were different. Obviously, the reason for the difference is that the oxygen contents for different crystals during their growing process are not exactly the same, so that the doping levels of carriers in the CuO_2 planes are different. However, we cannot distinguish from x-ray diffraction the slight differences in the crystal structures. The as-grown crystals in our experiments usually exhibit metallic conduction at high temperature but semiconducting-like behaviour at low temperature, i.e. a shallow minimum ρ_{min} appears in their ρ–T curves (see below). But the temperatures T_{min} corresponding to the resistivity minima ρ_{min} are dependent on the crystals. When the as-grown crystals were annealed in air or ambient flowing oxygen for a period, they could become metallic in the whole temperature range and even exhibit a superconducting transition at low temperature. By selecting the samples showing various temperature-dependent resistivities, we can study systematically the evolution of the charge conduction from metallic to insulating states. Because of the difficulty in measuring the oxygen content exactly, here we shall distinguish the doping levels in different crystals qualitatively by means of the relative resistivity and also of the temperature-dependent behaviours. Generally speaking, a sample has a higher resistivity if its T_{min} is higher. It is reasonable to believe that, the higher T_{min} the sample has, the less oxygen it contains, and thus the lower doping level it has.

Figure 1 shows the temperature dependence of the in-plane resistivity for three different crystals annealed in air. We can see that there is in general a trend of electron localization at low temperature. Crystal A is metallic in the whole temperature range with a superconducting transition near 4.2 K. A noticeable feature in its ρ–T curve is that the resistivity deviates from the linear temperature dependence and seems to cross over to a higher-power temperature dependence at temperature below 80 K. Though this type of ρ–T curve seems approximately to follow Bloch–Grüneisen formula, we suggest that the sample is in fact in the slightly underdoped region and the behaviour is caused most likely by the competition between the metallicity at high temperature and the tendency towards localization at low temperature [7]. The doping levels for crystals B and C are comparatively

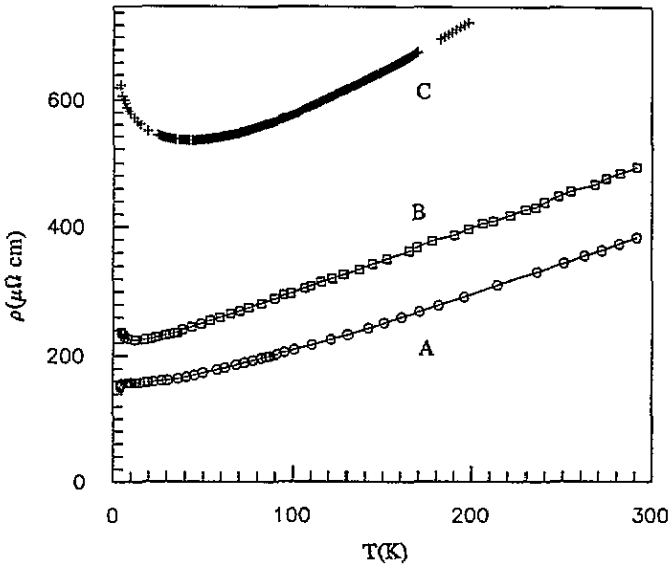

Figure 1. Temperature dependence of in-plane resistivity of samples A, B and C. Sample A shows a superconducting transition near 4.2 K. Localization behaviours were observed at low temperature for B and C.

lower than crystal A. They have higher resistivity and show distinctly localized conductions at low temperatures. The resistivity minimum ρ_{min} for crystal B appears at a temperature of about 12 K. For crystal C it is near 35 K. The resistivity follows approximately a linear temperature dependence at temperature higher than T_{min} in general.

The in-plane scattering mechanism in the normal state has been a central concern since the discovery of high-T_c cuprates. Important progress in the past two years has been to find some evidence for the charge transport being dominated by spin scattering. It is found that for underdoped $YBa_2Cu_3O_{7-y}$ crystals [8], $\rho_{ab}(T)$ deviates from T-linear behaviour and displays a downward curvature below a certain temperature but well above T_c. The deviation coincides with the development of a gap in the spin excitations seen in neutron and NMR studies. The suggestion is that in the region where the spin gap closes, at high temperature or at higher doping level, spin fluctuations would give rise to the T-linear in-plane resistivity. When the spin gap opens, the suppressed spin fluctuation would reduce in-plane scattering, leading to the decrease in ρ_{ab}. The ρ–T curve thus shows a downward curvature. A similar phenomenon was also observed in the $YBa_2Cu_4O_8$ crystal which belongs to the underdoped high-T_c superconductors [9]. Naturally, it is desirable to see if similar behaviour appears in other high-T_c cuprates. Our experiments suggest that no such transport feature associated with the 'spin gap' is present in Bi-2201 materials. Instead of a downward curvature in the ρ–T curve, we usually observed upward curvature. At present we have not learned of a detailed neutron scattering or NMR measurement on the Bi-2201 compound. We suggest two possible reasons for the difference. One might be that different systems have different spin excitation spectra. The other might be due to the strong localization tendency at low temperature, so that the feature associated with the spin gap is not observed in our measurement. This is the case in particular if the temperature of opening of a spin gap is near the superconducting transition temperature.

If we plot the temperature axis with a logarithmic scale (figure 2), we can see clearly that the resistivity below T_{min} for crystals B and C follows the relationship of $\rho(T) \sim \ln T$. This is the typical behaviour of a two-dimensional electronic system in the weak-localization regime [10]. We have also try to fit the low-temperature resistivity of sample C to the VRH formula $\rho = \rho_0 \exp(T_0/T)^\alpha$ ($\alpha = \frac{1}{4}$); though the fitting seems not bad, the value of T_0

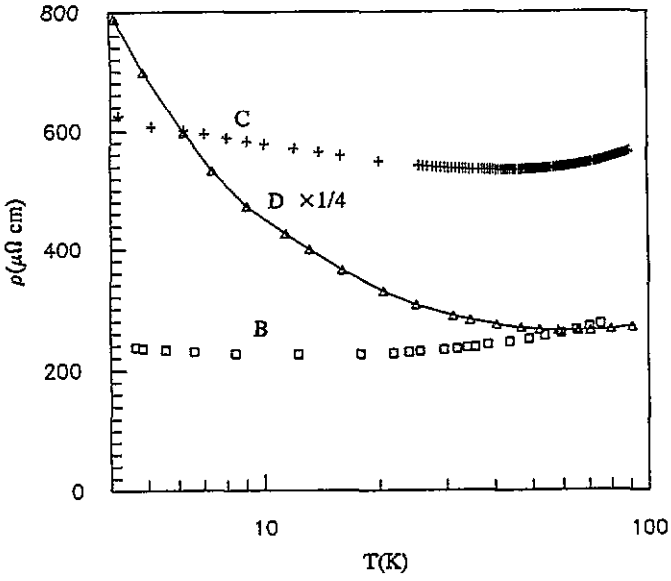

Figure 2. A plot of ρ–T curves for samples B, C and D in a semilogarithmic scale. Sample B and C at low temperatures follow the relationship of $\rho \propto \ln T$, suggesting two-dimensional weak-localization conductions. But sample D shows a faster than $-\ln T$ increase in resistivity as T goes down.

obtained is less than 1 K. One must keep in mind that hopping conduction requires the most probable hopping distance R_M being greater than the localization length ξ, i.e. $R_M/\xi > 1$. According to Mott's VRH theory [11], $T_0 \approx 16/k_B N(E_F)\xi^3$, $R_M = 3\xi(T_0/T)^{1/4}/8$. For the case of $T_0 < 1$, e.g. for crystal C in the fitting temperature range (4.2–15 K), one then gets $R_M/\xi \ll 1$, which is clearly in contrast to the assumption of VRH conduction.

The weak localization is due to the correction of the quantum interference effect of electron wavefunctions at the disorder impurities to the conductance, which results in the increase of the probability of electrons at the impurities. Applying an external magnetic field will suppress the quantum corrections and thus lead to a negative orbital magnetoresistance (MR) [10]. In order to see if this is the case, we have measured the MR of crystals in the weak-localization regime. A surprising feature is that a positive MR is generally observed at low temperature. Figure 3 is the transverse MR ($I \parallel ab$ plane, $H \parallel c$ axis) of the sample C. We can see positive MR appearing distinctly at low temperature. At a fixed temperature, the positive MR first increases with the magnetic field, then decreases slowly after reaching a maximum. The field H_{max} corresponding to the maximum positive MR shifts to lower field with increasing temperature. At high temperature (still in the weak-localization regime) and high field, negative MR is observed. From the shape of the MR curve, it is reasonable to consider that the appearance of the maximum in MR is caused by the competition between the positive MR appearing at low temperature and low field and the negative MR appearing at high temperature and high field. As negative MR should exist in the weak-localization region, the main focus is the origin of the positive MR at low temperature. It is most likely that the positive MR is contributed from superconducting fluctuations, because very strongly positive MR appears in the superconducting fluctuation regime. It increases first with the field, and soon becomes saturated. Therefore our experiment suggests that crystal C is near the border of the superconducting–insulating transition. It might be superconducting at very low temperature. However, due to the complex contributions from both the superconducting fluctuation and weak-localization effect, we cannot derive the temperature dependence of the inelastic scattering relaxation rate from the MR measurement.

Figure 4 shows the temperature dependence of the in-plane resistivity for three as-grown

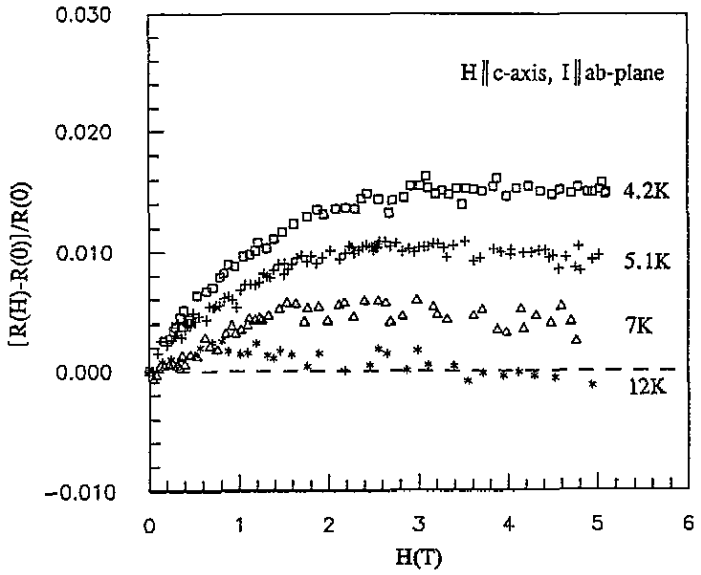

Figure 3. The transverse MR of crystal C at different temperatures.

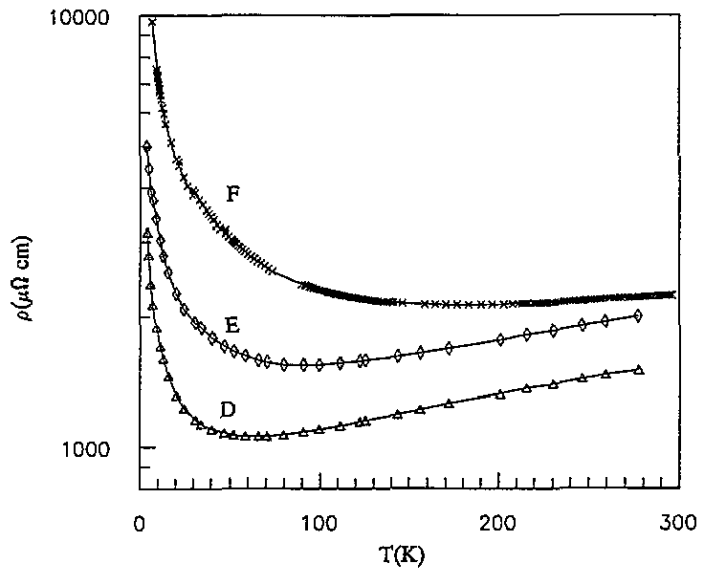

Figure 4. Temperature dependence of in-plane resistivity of three as-grown samples D, E and F. They show stronger localization behaviours at low T.

crystals. Strongly localized behaviours appear at low temperatures. We also plot the ρ–T curve for sample D with semilogarithmic coordinates; it shows a faster than $-\ln T$ increase in resistivity below T_{min} with the decrease of temperature (figure 2). This indicates definitely that the crystal is in the strong-localization regime at low temperature. We then fit the $\rho - T$ curve with the VRH theory of Mott. In figure 5, the data of crystals D and F are plotted as $\ln \rho$ against $T^{-\alpha}$ with $\alpha = \frac{1}{4}$ to show the hopping behaviour. We then get $T_0 \approx 172$ K for sample D and $T_0 = 477$ K for sample F. According to Mott's theory, the localization length ξ of sample F is shorter than that of sample D, which implies that sample F has a even stronger localization effect. In fact, sample D as well as sample F are in a critical regime where the VRH theory can be used due to the small values of T_0.

As we mentioned at the beginning VRH conduction has been found in various

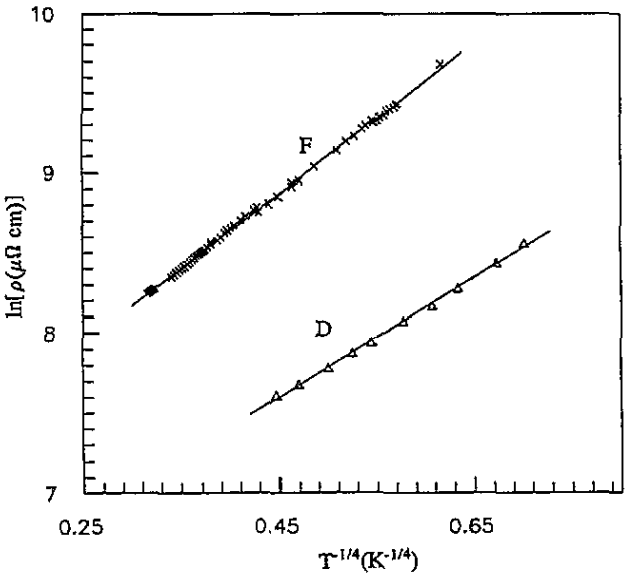

Figure 5. Variable-range hopping $\rho = \rho_0 \exp(T_0/T)^{1/4}$ fits to the low-temperature resistivity of crystals *D* and F.

high-T_c cuprate systems, for example, in $La_{2-x}Sr_xCuO_y$, [3] $Y_{1-x}Pr_xBa_2Cu_3O_7$ [12], $Bi_2Sr_2Ca_{1-x}Y_xCu_2O_y$ [6]. It is often found that, in these systems, the fitting exponent α increases from $\frac{1}{4}$ (or even $\frac{1}{5}$) to $\frac{1}{2}$ with the decrease of doping levels. Fiory *et al* [13] reported that a best fit of the VRH formula to their data of Bi-2201 crystals required $\alpha = \frac{1}{3}$. We believe that the crossover from two-dimensional weak localization to hopping conduction and further of the exponent α from $\frac{1}{4}$ (or $\frac{1}{5}$) to $\frac{1}{2}$ with the decrease of doping is a universal trend for these cuprates, and reflects the revolution of the electronic behaviours under the process of M–I transition. Because our samples are in the vicinity of the M–I transition, we therefore fitted the data to the VRH formula with $\alpha = \frac{1}{4}$ (figure 5). In fact, a fitting of the data of sample F to $\alpha = \frac{1}{3}$ appeared not so good as that to $\alpha = \frac{1}{4}$. But here we shall not address the question of dimensionality from the fitting in the VRH regime because the theoretical assumptions used in deriving the exponents $\alpha = \frac{1}{3}$ and $\frac{1}{4}$ may not be fulfilled in the layered cuprates [6].

We can see that the M–I transition here takes the form of an Anderson transition, i.e. electrons being localized from the weak regime to the strong regime. The physical origin for Anderson localization is the disordering effect of (impurity) potentials. Under the weak-localization regime, the disordering effect is weak and the mean free path of electrons l is much greater than the electron wavelength $\lambda = 2\pi/k_F$. Though the disordering at impurities cannot bond the electrons, one must take into account the correction of the quantum interference effects to the conductance. For two-dimensional system, both the scaling theory and the many-body theory give the relation of $\Delta\sigma \propto \ln T$. With the increase of disordering, l becomes shorter. As the disordering effect is so strong that $l \approx \lambda$, the wavefunction of the electron decays exponentially, i.e. $|\Psi(r)| \propto e^{-|r|/\xi}$, which means the electron is localized at an impurity with the range length of ξ. This is the case of strong localization, in which the electron conduction with a VRH formula was established. What we have observed in our experiment is just the crossover behaviour from a weak-localization regime with a logarithmic scaling to a strong-localization regime with an exponential scaling.

However a simple analysis suggests that the disordering of the lattice does not become more prominent with the doping level decreasing. We can see this point from a simple

example of the $La_{2-x}Sr_xCuO_y$ compound. Definitely with the reduction of Sr content the doping level is reduced in this system, and accompanying this the M–I transition takes place. But it is certain that the crystal lattice becomes more ordered with the reduction of substitutions of Sr for La sites. In general, the in-plane Cu–O bond is very strong in the lattice, and should not be affected largely by the out-of-plane changes, like element substitutions and oxygen vacancies. In other words, the effects of lattice imperfections in other layers on the conduction of electrons in CuO_2 layers should be in a secondary position. This statement is further strengthened by the fact that the optimal doping high-T_c cuprates exhibit almost a zero residual resistance and nearly the same slope in ρ–T curves [14]. The above observation and analysis leads us to explain why the effect of disordering (not the disordering itself) becomes stronger and stronger with the reduction of carrier density.

Our suggestion is that the electron correlation plays an important role. Both the correlation and disordering are present in high-T_c cuprates and enhance each other's effect. In a general view, the conduction of an electron is determined by its kinetic energy and the potentials (interactions). The M–I transition takes place when the kinetic energy of the electron is not large enough to overcome the potential energy. It is well known that the parent compound of high-T_c cuprates is a half-filling charge transfer insulator with a charge transfer energy gap of about 1.5–2 eV. This is the case due to the strong on-site (Cu) electron correlation, which is well understood in the framework of the Hubbard model in the large-U limit. Upon doping, the system deviates from half-filling, and mobile carriers are created mainly with O 2p character. Physically the system should be described by a three-band Hubbard model with doping, where the interactions between holes at the same d, p orbitals as well as at adjacent Cu–O d–p orbitals are important parameters [15]. On one hand, even if there were no random potentials, these interactions (or effective interactions) would get stronger as the screening of electron became poor due to the reducing of doping. Relatively the kinetic energy of the electron might become comparable with or even smaller relative to the potentials. As a consequence, it is unfavourable energetically for the electron to move. In the case of disordering, the random potentials would become dominant. That is the localization effect we have observed. On the other hand, as the Anderson transition is approached, the carrier diffusion slows down and in effect they interact more strongly [1].

However one should note that the mechanisms for the M–I transition in different strongly correlated electron systems are not the same. They depend on the materials. A noteable example is the $La_{1-x}Sr_xTiO_3$ system [16–18], which became of particular interest after the discovery of high-T_c superconductivity. This is because the parent compound $LaTiO_3$ is a Mott–Hubbard insulator, with antiferromagnetic ordering of Ti^{3+} spins (d^1 configuration with spin $= \frac{1}{2}$, similar to the parent compounds of high-T_c cuprates) at $T_N = 120$–150 K. Substituting La with Sr, the compounds become metallic abruptly. In contrast to high-T_c cuprates the metallic state follows the description of Landau's theory of a Fermi liquid, and the mobile carriers, which are electrons (not holes), decrease distinctly with the increase of the Sr content. Analyses on the measurements of optical conductivity and specific heat of this system indicate that the M–I transition here is caused by the divergence of the effective electron mass ($m^* \rightarrow \infty$). This is very different from cuprates, in which the M–I transition is driven by decrease in the carrier concentrations. It deserves more attention that the M–I transition in a doped two-dimensional charge transfer insulator is different from that in a doped three-dimensional Mott–Hubbard insulator.

In conclusion, we have concentrated our attention on the localized behaviours in $Bi_2Sr_2CuO_y$ crystals. An evolution of electronic states from a weak-localization regime to a strong-localization regime with the reduction of doping level was presented and discussed.

We conclude that the electron localization and also the M–I transition occurring in high-T_c cuprates are driven by the reduction of carrier concentration. In the process of the transition the electron correlation and disorder are present together and enhance each other's effects.

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