Unconventional Transition from Metallic to Insulating Resistivity in the Spin-ladder Compound (Sr,Ca)$_{14}$Cu$_{24}$O$_{41}$

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Spin-ladder compounds make interesting analogs of the high-temperature superconductors [1], because they contain layers of nearly one-dimensional “ladders” consisting of a square array of copper and oxygen atoms. Increasing the number of legs in the ladders provides a step-wise approach toward the two-dimensional copper-oxygen plane, that structure believed to be a key to high temperature superconductivity. Short-range spin correlations in ladders have been predicted to lead to formation of hole pairs favorable for superconductivity, once enough holes are introduced onto the ladders by doping [2]. Indeed, superconductivity has been discovered in the two-leg ladder compound (Sr,Ca)$_{14}$Cu$_{24}$O$_{41}$ under high pressure [3]. Here we show that charge transport in the non-superconducting state of (Sr,Ca)$_{14}$Cu$_{24}$O$_{41}$ shares three distinct regimes in common with high-temperature superconductors, including an unexplained insulating behavior at low temperatures in which the resistivity increases as the logarithm of the temperature. These observations suggest that the logarithmic divergence arises from a new localization mechanism common to the ladder compounds and the high-temperature superconductors [4], which may arise from nearly one-dimensional charge transport in the presence of a spin gap.

It is increasingly believed that a successful theory of high-temperature superconductivity (HTS) must also account for the anomalous properties exhibited by the so-called “normal state”, the resistive state observed at temperatures above the superconducting transition temperature, $T_c$. There are numerous similarities in the normal-state properties of HTS and ladder cuprates, including an energy gap for spin excitations and a crossover from insulator to metal upon hole doping. These suggest that the ladder compounds provide a valuable experimental laboratory for probing the unusual properties of the resistive normal state of the high-temperature superconductors.

In this letter we discuss resistivity measurements on five single-crystal samples of Sr$_2$Ca$_{12}$Cu$_{24}$O$_{41}$, which were grown by the traveling-solvent-floating-zone method [5]. As shown in Fig. 1, the crystal structure of this compound is composed of layers of CuO$_2$ chains and Cu$_2$O$_3$ two-leg ladders, interleaved with Sr$_{1-x}$Ca$_x$ buffer layers. The undoped parent compound, Sr$_{14}$Cu$_{24}$O$_{41}$, exhibits semiconducting behavior with an activation energy gap of 0.18eV [5]. The formal valency of Cu in Sr$_{14}$Cu$_{24}$O$_{41}$ is +2.25, corresponding to a partially-filled valence band which ordinarily would result in metallic conductivity. However, in Sr$_{14}$Cu$_{24}$O$_{41}$, the positively-charged carriers (the “holes”) are located on the CuO$_2$ chains, which do not conduct because the transfer integral along the 90° oxygen bond is small (Fig. 1(b)). Upon partial substitution of Ca for the isovalent Sr (or upon the application of pressure), the holes are redistributed from the chains to the ladders, which are more conductive due to the 180° oxygen bonds [5]. This “self-doping” leads to a decrease of resistivity and, eventually, a crossover from insulating to metallic behavior as the carrier concentration is increased.

At lower temperatures, both samples experience a crossover from metallic to insulating behavior ($d\rho_c/dT < 0$). Although both samples have the same nominal composition, the insulating behavior begins at a higher temperature in sample “E” and the low-temperature resistivity is roughly two orders of magnitude larger in sample “E” than in sample “D”. This difference can be due to different amounts of disorder and/or different levels of the carrier concentration between two samples. Nevertheless, in the extreme low-temperature limit, $\rho_c$ for both samples follows the temperature dependence expected for variable range hopping (VRH) of strongly localized carriers: $\rho_c = \rho_0 \exp(T_0/T)\beta$. The best fit to the data over the widest temperature range (Fig. 2) corresponds to $\beta = 1/2$, the same temperature dependence reported in highly resistive samples of the HTS cuprates [6,7]. In this VRH regime, electrical current is dominated by variable-length hops of the charge carriers between localized states at the minima of the random disorder potential. The $\beta = 1/2$ exponent can result from VRH in the presence of Coulomb repulsion between carriers, which suppresses the density of states at the Fermi energy in a low-carrier-density system [8].

The most surprising feature of the data occurs in an...
intermediate temperature regime between 3K and 20K in sample “D” (Fig. 3). This regime is characterized by a temperature dependence best approximated by a logarithmic insulating behavior, $\rho_c \sim \log(1/T)$ \[13\]. In this log-$T$ regime, the magnetoresistance (MR) of sample “D” (measured at $T = 4.2$K with the magnetic field applied along the b-axis) was found to be negative, while in the strong localization regime (measured at $T = 1.2$K) the MR was found to be positive, as is typical for VRH. This gives additional experimental evidence that there is a third transport regime in sample “D”, distinct from the linear-$T$ and strong localization regimes.

Of the three transport regimes, the lowest-temperature strong localization regime is the most robust, since VRH is observed at temperatures below 2K in every sample. This is the same temperature range in which long range spin order has been reported in (Sr,Ca)$_{14}$Cu$_{24}$O$_{41}$, in which spins on neighboring Cu-ladder sites are anti-aligned \[14\]. It is consistent with our observations to suppose that the onset of long range anti-ferromagnetic order gives rise to the onset of strong localization of charge carriers in the ladders.

The other two transport regimes observed in sample “D” are not nearly so robust, perhaps signaling a greater sensitivity to disorder for the underlying transport mechanisms. After all, quasi-1D transport would be particularly sensitive to disorder: a mobile charge on a ladder may well have difficulty getting past a defect or break in that ladder. The cleanest linear-$T$ dependence is observed in sample “D”, while samples with higher room-temperature resistivity tend to show quasi-linear dependence of $\rho_c$ for which the extrapolated zero-temperature resistivity is non-zero. In samples with sufficiently high room-temperature resistivity (> 2m$\Omega$ cm), insulating behavior ($d\rho_c/dT < 0$) is observed even at room temperature, probably due to larger amounts of disorder in these samples. Although evidence of the log-$T$ regime has been found in two samples, we note that the unambiguous observation of the log-$T$ regime occurs in the sample which exhibits the cleanest linear-$T$ regime ($\rho_c \sim aT$) of all samples studied.

Like the linear-$T$ transport regime, a log-$T$ transport regime also exists in the normal state of the HTS cuprates. There is growing experimental evidence that the anomalous normal-state properties of the HTS are due to the strong electron interactions near an insulator-to-metal crossover. The crossover is ordinarily obscured in HTS by the appearance of the superconducting phase; however, by suppressing superconductivity with an intense, pulsed magnetic field, the insulator-to-metal crossover in La$_{2-x}$Sr$_x$CuO$_4$ has been found to occur near optimum doping \[15\], that carrier density which yields the maximum $T_c$. Underdoped samples, those with fewer...
carriers than optimal doping, exhibit a transport regime characterized by a log-\(T\) divergence of the normal state resistivity, once superconductivity is lifted by the magnetic field\(^\text{[4,5]}\). This divergence is inconsistent with known models for log-\(T\) insulating behavior: it is apparently not arising from weak localization due to coherent backscattering\(^\text{[4,5]}\) or disorder-enhanced electron interactions, neither is it likely due to spin-flip (Kondo) scattering\(^\text{[4,5]}\).

While there are differences between the CuO\(_2\) plane of HTS cuprates and the plane containing 2-leg ladders in Sr\(_2\)Ca\(_2\)Cu\(_{24}\)O\(_{41}\), we note that the log-\(T\) insulating behavior occurs in both systems at the same magnitude of normalized resistivity, when the resistivity per layer is near the quantum resistance, \(h/e^2 \sim 25.8\, \text{k}\Omega\). In a conventional two-dimensional system the quantum resistance corresponds to that resistance at which the mean free path is comparable to the deBroglie wavelength at the Fermi energy, which is where transport typically crosses from metallic (diffusive) to insulating (localized) behavior. It is tempting to suggest that similar physical mechanisms can govern normal-state transport properties in the two-leg ladder compound and the HTS cuprates, even though, prima facia, the former contains quasi-one-dimensional transport along Cu\(_2\)O\(_3\) ladders, while the HTS cuprates contain quasi-two-dimensional transport in the CuO\(_2\) plane. Nonetheless, the phenomenology of three different transport regimes is similar in the two systems, including the two regimes (linear-\(T\) and log-\(T\)) for which the underlying physical mechanisms remain unknown. In light of the data presented here, when coupled with published evidence of charged-stripe formation in HTS\(^\text{[17]}\), one could speculate that the effective dimensionality of the HTS CuO\(_2\) plane is reduced with regard to charge transport and that the charge-transport mechanisms are similar in the two systems.

FIG. 3. Variable range hopping (dashed lines) in the lowest temperature range for both samples.

FIG. 4. Intermediate temperature regime (3K < \(T\) < 20K) in sample “D”, which is best described by \(\rho_c(T) \sim \log(1/T)\). Dashed line is the extrapolation of the VRH fit from Fig. 3. Right axis shows normalized \(\rho_c\) per layer of Cu\(_2\)O\(_3\) ladders, in units of the quantum resistance, \(h/e^2\).

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We note that the data in Fig. 4 has a slight curvature in the semi-log plot suggesting a dependence somewhat weaker than purely logarithmic. This curvature is reproducible and larger than any known measurement errors.

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