Magneto-transport in LSCO high-$T_c$ superconducting thin films

Fedor F Balakirev$^1$, Jon Betts$^1$, Gregory S Boebinger$^2$, I Tsukada$^3$ and Yoichi Ando$^3$

$^1$ National High Magnetic Field Laboratory, Los Alamos National Laboratory
Los Alamos, NM 87545, USA
$^2$ National High Magnetic Field Laboratory, Florida State University,
Tallahassee, FL 32310, USA
$^3$ Central Research Institute of the Electric Power Industry, Tokyo, Japan
E-mail: gsb@magnet.fsu.edu

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Abstract. We report a crossover from insulating to metallic behaviour near optimal doping in high-$T_c$ thin films of La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) using pulsed magnetic fields to suppress superconductivity and reveal the normal state magneto-transport in the zero temperature limits. The thin film data supports the same key findings reported on LSCO single crystals: (i) that the crossover from insulating to metallic behaviour occurs near optimal doping, and (ii) that the insulating behaviour is characterized by an unusual logarithmic divergence of the resistivity as the temperature approaches zero. A new finding is that the Hall density exhibits a sudden increase at the same Sr doping level at which the insulator-to-metal crossover occurs, possibly indicating a sharp change in the Fermi surface at optimum doping.
1. Introduction

After almost two decades since the discovery of high temperature superconductors (HTS), the origin of high temperature superconductivity remains a central controversy in condensed matter physics. Even the phase diagram is still the subject of active debate, and many believe that the clearly unconventional nature of the normal state of the high-$T_c$ superconductors may well result from the same physics that underlies high temperature superconductivity. The biggest obstacle to observations of the ground state in the HTS is the superconductivity itself, which effectively conceals the properties of the normal state at low temperatures. A number of models for HTS are based on the concept that both HTS and the anomalous electronic properties of the normal state arise from competition between still-uncharacterized ground states. Extremely high magnetic fields provide an unprecedented opportunity to suppress superconductivity to reveal signatures of what appears to be a quantum phase transition in the normal state of HTS in the zero-temperature limit.

The two HTS La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) and Bi$_2$Sr$_{2-x}$La$_x$CuO$_{6+\delta}$ (BSLCO) have been the main focus of magneto-transport measurements in high magnetic fields at low temperatures. Non-destructive pulsed magnets now routinely provide magnetic fields of 60–65 T. A field of order 60 T is required to completely suppress superconductivity in these two HTS compounds, the two that exhibit a maximum superconducting transition temperature $T_c \leq$ 40 K at optimum doping. Still higher magnetic fields are required to suppress superconductivity in HTS compounds with higher maximum $T_c$.

The unit cells of LSCO and BSLCO contain a single CuO$_2$ plane, the key structural element that is common to all HTS. High quality LSCO and BSLCO single crystals can be reliably produced over a wide range of carrier concentrations by the inter-substitution of Sr and La, an important consideration for systematic doping-dependent studies. The accessible hole doping range spans the underdoped Mott insulator limit, through the superconducting phase, to the overdoped regime.

To date, almost all knowledge of the properties of the normal state of HTS at low temperatures has been obtained from magneto-transport measurements. One of the most striking results is the discovery of the insulator-to-metal crossover underlying the superconducting phase in both LSCO [1] and BSLCO [2] single crystals, once the superconducting phase is suppressed with high magnetic fields. The insulating behaviour of the underdoped superconductors is found to follow an anomalous logarithmic divergence of resistivity at low temperatures, the origin of which remains unresolved [3]. Recent Hall voltage measurements of BSLCO single crystal reveal a cusp in the normal state Hall number at optimum doping [4]. The data suggest the presence of a zero-temperature phase transition precisely at the same carrier concentration at which high
temperature superconductivity is most stable. A primary motivation for these thin film LSCO experiments has been to ultimately make a detailed study of the Hall resistivity throughout the temperature-doping phase diagram for a second HTS compound.

2. Experiments

Measurements in the low temperature ground state in several of the most common HTS compounds are made possible due to recent advances in high magnetic field technology. The 60 T high magnetic field provides a unique capability to suppress superconductivity without changing normal state behaviour; however, it also imposes relatively strict constraints, including limited experimental volume (~5 mm × 5 mm × 5 mm at peak field) and only a few milliseconds at peak magnetic field for each magnet pulse. Increased noise background results from the rapid time-varying magnetic field that swings to 60 T and back to zero within ~100 ms, during which time the sample is positioned within millimetres of the magnet windings that carry tens of kilo-amperes, experience kilovolt potential drops between neighbouring windings, and likely move in response to the mechanical stresses resulting from the Lorentz forces experienced. To overcome these difficulties, we developed a customized synchronous digital lock-in technique, designed and developed at the National High Magnetic Field Laboratory to acquire all information produced by a measurement in the shortest time permitted by information theory, with minimum sensitivity to noise and electrical interference. The dual frequency signal/clock function generator and advanced lock-in software algorithms realize sub-microvolt signal detection at frequencies up to a megahertz with synchronous time constants as short as one cycle of the sine wave excitation. Most of the data in this paper were measured using this technique.

The LSCO thin film samples were prepared by laser ablation using strontium titanate (STO) substrates [5] with eight different levels of Sr doping spanning the range of \( x \) from 0.08 to 0.22. The superconducting state in LSCO results from doping the Cu-O planes with holes via partial substitution of La with Sr. All samples show metallic behaviour, i.e. decreasing resistivity as temperature is decreased, at all temperatures above \( T_c \). The highest \( T_c \) is achieved at Sr concentration \( x = 0.17 \). The samples were patterned in a conventional Hall bar geometry to facilitate accurate resistivity and Hall measurements, with magnetic field applied perpendicular to the film.

3. Results and discussion

Shown on figure 1 is a typical example of magnetic field dependencies of resistivity at several different temperatures for two thin film samples \( \text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4 \) (underdoped) and \( \text{La}_{1.8}\text{Sr}_{0.2}\text{CuO}_4 \) (overdoped). The resistivity shows minimal change with magnetic field for temperatures above \( T_c \). Upon cooling down below \( T_c \), there is a distinct superconducting phase with zero resistivity in the low field region. As the magnetic field increases, superconductivity is eventually suppressed until the resistive response becomes that of the normal state in the absence of superconductivity. The underdoped sample \( \text{La}_{1.86}\text{Sr}_{0.14}\text{CuO}_4 \) shown on figure 1(a) displays a clear insulating behaviour at low temperatures—the value of the normal state resistivity observed at high magnetic field increases as the temperature decreases, while the resistivity in the overdoped sample \( \text{La}_{1.8}\text{Sr}_{0.2}\text{CuO}_4 \) shown on figure 1(b) is metallic. This transition from an insulating to a metallic normal state in otherwise superconducting samples has been first reported for single crystals of LSCO [1].

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Figure 1. Selected traces of in-plane resistivity $\rho_{ab}$ versus magnetic field (a) in La$_{1.86}$Sr$_{0.14}$CuO$_4$ thin film and (b) in a La$_{1.8}$Sr$_{0.2}$CuO$_4$ thin film.

Figure 2. Temperature dependence of the in-plane resistivity for LSCO thin films in zero magnetic field (solid lines) and in pulsed magnetic fields of 50 T (symbols) at several different hole doping levels, $x$. Note that the insulating behaviour crosses to metallic behaviour at $x \sim 0.195$. The separatrix at $\rho_{ab} \sim 0.09$ m$\Omega$ cm corresponds to $k_F\ell \sim 18$, well on the metallic side of the Mott–Ioffe–Regel limit. The left-hand side panel is enlarged to show the insulator to metal crossover more clearly.

The fact that application of high magnetic fields is required to uncover the transition from metallic to insulating ground state is clearly demonstrated in figure 2. This figure shows the temperature dependence of resistivity in the subset of LSCO thin films near optimum doping, with $0.14 < x < 0.22$. Both the zero field resistivity (solid lines) and the resistivity at 50 T (symbols) are shown. While all the samples shown in figure 2 are metallic at zero field above $T_c$, the 50T magnetic field suppresses superconductivity and reveals that all samples up to $x \sim 0.19$ doping show insulating behaviour at low temperatures. The low temperature separatrix between
insulating and metallic behaviour occurs at a resistivity of $\sim 0.09 \, \text{m} \Omega \, \text{cm}$, which corresponds to $k_F \ell \sim 18$, where $k_F$ is the Fermi wave vector and $\ell$ is the mean free path. This value is strikingly similar to that found in LSCO single crystals: $k_F \ell \sim 15$ [1]. In addition, the suddenness of the insulator to metal crossover at $x \sim 0.195$, with $x = 0.19$ showing insulating behaviour and $x = 0.20$ showing metallic behaviour, reproduces behaviour seen in LSCO single crystals, which show the same phenomena, with the insulator to metal crossover occurring at $x \sim 0.16$ in the single crystal samples studied [1]. It is an important observation that our LSCO films reproduce the results from LSCO single crystals. It is worth noting that LSCO thin film results do not necessarily reproduce the behaviour of LSCO single crystals. Whereas we report insulating behaviour throughout the underdoped regime for LSCO films grown on STO, this behaviour is not seen in LSCO films grown on SrLaAlO$_4$, which imparts a compressive in-plane strain on the LSCO film. In a compressively-strained LSCO film with $x \sim 0.10$, $T_c$ is enhanced and it has been reported that metallic behaviour replaces the $\log(1/T)$ resistivity divergence seen in our films and in single crystals [6].

We turn our attention to Hall measurements, which on BSLCO single crystals are facilitated by the thin ($\sim 10 \, \mu\text{m}$ thick) platelet geometry in which BSLCO samples naturally grow. Accurate Hall measurements in the restrictive environment of pulsed magnetic fields require thin samples, because the Hall voltage is extremely small and inversely proportional to the thickness of the sample. LSCO single crystals grow in thick crystals that resist cleaving and mechanical thinning of LSCO has proven inadequate for pulsed magnetic field measurements.

Figure 3 shows the typical field dependence of the Hall resistivity in superconducting LSCO thin films. The magnetic field suppresses superconductivity even at the lowest temperatures...
Figure 4. Doping dependence of the Hall density (defined as $1/R_H$) in thin films of LSCO above the superconducting transition temperature. Note the relatively abrupt increase in Hall number above Sr doping, $x \sim 0.17$. The region in black is the superconducting phase, in which the Hall number is zero.

shown. The signal-to-noise in these thin film LSCO measurements is sufficiently high that we can conclude that the low-temperature ($T < T_c$) Hall signal recovers its conventional linear-in-magnetic-field behaviour, once superconductivity is suppressed by the high magnetic field. The value of the Hall coefficient in the normal state, $R_H$, is then determined from a linear fit to the Hall resistivity data in this high field regime, $\rho_H(H) = R_H H$ (dashed lines in figure 3).

Figure 4 shows the temperature and doping dependence of the Hall density, $1/R_H$, in the normal state of LSCO. We find that the Hall number remains relatively low in underdoped samples and, above $x \sim 0.17$ the Hall number rather suddenly assumes a much faster increase with an increase in doping. This behaviour, as well as the abrupt crossover from insulating to metallic behaviour in the $\rho_{ab}$ data of figure 2, is not seen in BSLCO, in which both transitions appear much more gradually as doping is increased. Eventually, the data in figure 4 will diverge as the Fermi surface of LSCO changes from ‘hole-like’ to ‘electron-like’. In this regard, for $x > 0.20$, where the Hall number greatly exceeds the Luttinger volume of $1 + x$ holes per copper, the data are evidencing the eventual change in sign of the Hall voltage above $x = 0.30$ [5].

4. Summary

Our observations from LSCO films are qualitatively consistent with a change in the Fermi surface near optimum doping, from a low density metal to a higher density metal. Recent active debate has centred around a number of models based upon the existence of a quantum phase transition between two different metallic phases in the normal state of the HTS. It takes an extremely high magnetic field to suppress the superconductivity and reveal this regime near optimum doping,
but data on LSCO single crystals, BSLCO single crystals, and now LSCO thin films are all consistent with the still-qualitative nature of these theoretical models.

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