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History dependent magnetoresistance in lightly doped $La_{2-x}Sr_xCuO_4$ thin films

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ABSTRACT

The in-plane magnetoresistance (MR) in atomically smooth $La_{2-x}Sr_xCuO_4$ thin films grown by molecular-beam-epitaxy was measured in magnetic fields B up to 9 T over a wide range of temperatures T. The films, with $x=0.03$ and $x=0.05$, are insulating, and the positive MR emerges at $T<4$ K. The positive MR exhibits glassy features, including history dependence and memory, for all orientations of B. The results show that this behavior, which reflects the onset of glassiness in the dynamics of doped holes, is a robust feature of the insulating state.

1. Introduction

Understanding the origin and the role of various nanoscale inhomogeneities observed in underdoped cuprates is one of the major open issues in the study of high-temperature superconductivity (HTSC). The nature of the ground state at low charge carrier concentrations is of particular interest, because it is from this state that HTSC emerges with doping. While the long-range antiferromagnetic (AF) order of the parent compound is already destroyed at very low level of hole doping (*e.g.* $x\approx 0.02$ in $La_{2-x}Sr_xCuO_4$), short range AF correlations of the Cu spins persist [1]. In $La_{2-x}Sr_xCuO_4$ (LSCO), each of these AF domains has a weak ferromagnetic (FM) moment associated with it and oriented along the c axis, *i.e.* perpendicular to CuO_2 (ab) planes. The direction of the FM moment is uniquely linked to the phase of the AF order [2–4]. At a relatively low temperature T_{SG} (T_{SG}—spin glass transition temperature), the moments in different AF domains undergo cooperative freezing [5], so that the ground state of Cu spins is the so-called cluster spin glass (SG). The SG phase in LSCO emerges with the first added holes and extends all the way to slightly overdoped $x\simeq 0.19$ [6,7]. On the other hand, several experiments suggest that, in lightly doped (nonsuperconducting) LSCO, charge is clustered in antiphase boundaries [4,8–10] that separate the hole-poor AF domains in CuO_2 planes [11–15]. The nature of the charge ground state, however, has only recently attracted more attention.

In particular, in LSCO with $x=0.03$, which does not superconduct at any T, resistance noise spectroscopy [16,17] shows that, deep within the SG phase ($T\ll T_{SG}$), doped holes form a collective, glassy state of charge domains or clusters located in CuO_2 planes. The results strongly suggest that glassy freezing of charges occurs as $T\to 0$. These conclusions are supported by impedance spectroscopy [18]. In the same T range, both out-of-plane and in-plane magnetoresistance (MR) exhibit the emergence, at low fields, of a strong, positive component for all orientations of the magnetic field B [16,19]. The positive MR (pMR) grows rapidly with decreasing T and thus dominates the MR in the entire experimental B range at the lowest T. At higher T and B, on the other hand, the MR is negative. The mechanism of the negative MR at high T is attributed [20] to the reorientation of the weak FM moments. Most strikingly, unlike the negative MR, the pMR reveals clear signatures of glassiness, such as hysteresis and memory [16,19,21]. Similar behavior was observed in $La_2Cu_{1-x}Li_xO_4$ (Li–LCO) with $x=0.03$, where long-range AF order is still present [19]. This material, however, remains insulating for all x [22], and dielectric response provides evidence for slow [18,23] and glassy [23] charge dynamics in Li–LCO at low x. Detailed studies of the hysteretic and memory effects in the pMR of single crystals of both $La_{1.97}Sr_{0.03}CuO_4$ [16,17,19,21] and $La_2Cu_{0.97}Li_{0.03}O_4$ [19,24] have provided strong evidence that such history dependent behavior reflects primarily the dynamics of doped holes.

It is clearly of great interest to investigate the evolution of this glassy charge state with doping. For that purpose, LSCO films grown by molecular beam epitaxy (MBE) are particularly suitable because, in addition to their uniform thickness and precise crystal

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orientation, the doping can be controlled continuously and with high accuracy. Obviously, it is necessary to establish first whether history dependent positive MR observed in single crystals is also present in MBE-grown films, and thus independent of the growth conditions. Here we present a study of the low-T magnetotransport in MBE-grown LSCO films with $x=0.03$ and $x=0.05$, which demonstrates the presence of the pMR and the associated glassy effects.

2. Experiment

The LSCO films were grown by atomic-layer-by-layer molecular beam epitaxy (ALL-MBE) [25] on $LaSrAlO_4$ substrates with the c axis perpendicular to the surface. The films were deposited at $T \approx 680$ °C under 3×10^{-6} Torr ozone partial pressure. The growth was monitored in real-time by reflection high energy electron diffraction (RHEED), which showed that the films were atomically smooth and without any secondary-phase precipitates. The films are 75 unit cells (about 100 nm) thick; the measured $c=1.312$ nm. Finally, 160 nm of gold was evaporated *in situ* on top of the films for contacts. The films were patterned using UV photolithography and ion milling to fabricate Hall bar patterns with the length $L=2$ mm and the width $W=0.3$ mm. The distance between the voltage contacts is 1.01 mm, and their width is 50 μm. In order to remove any excess oxygen, the films were subsequently annealed in high vacuum ($4-5 \times 10^{-5}$ mbar) for over an hour at 200−250 °C.

The in-plane sample resistance R was measured with a standard four-probe ac method (∼11 Hz) in the Ohmic regime, at T down to 0.3 K realized in a ^{3}He cryostat. In the MR measurements, the current $I \perp B$.

3. Results and discussion

Both samples exhibit insulating behavior, with a two-dimensional (2D) form of the variable-range hopping (VRH) $R=R_0 \exp(T_0/T)^{1/3}$ obeyed up to about 70 K in $La_{1.97}Sr_{0.03}CuO_4$ (3% LSCO) and 40 K in $La_{1.95}Sr_{0.05}CuO_4$ (5% LSCO), as shown in Fig. 1. The localization length ξ obtained from the VRH fits [26] is ∼40 Å in 3% ($T_0=597$ K) and ∼60 Å in 5% ($T_0=244$ K) LSCO. Thus the doped holes are here localized more strongly than in a high-quality LSCO single crystal [28] with a nominal doping of 3%, where $\xi \sim 90$ Å and 2D VRH is obeyed up to 30 K [17,19,21]. In both cases, the range of the data is sufficiently wide to allow a reliable determination of the VRH exponent and without the need to invoke a T dependence of the prefactor R_0. The exponent 1/3, characteristic of 2D hopping, does not depend on doping, in contrast to early results on ceramic LSCO samples [29], but in analogy with more recent studies of polycrystalline LSCO [30]. Since the VRH conduction in both LSCO films and single crystals is dominated by 2D physics, at least in the experimental T range (see also Ref. [20]), the thickness of the films is not expected to affect the dimensionality of the transport properties.

Similar to the behavior in $x=0.03$ $La_{2-x}Sr_xCuO_4$ and $La_2Cu_{1-x}Li_xO_4$ single crystals [16,19], at low enough T, the MR of both 3% and 5% LSCO films exhibits the emergence of the positive component at low magnetic fields B for all field orientations. Here the pMR appears for $T<3-4$ K, as illustrated in Fig. 2(a) for 3% LSCO with B applied parallel to the c axis. As T is reduced, the pMR increases in magnitude and dominates the MR over an increasingly large range of B (Fig. 2(b)). History dependent behavior is present only in the B region where the pMR was initially observed (Fig. 2) after zero-field cooling. If the applied B is sufficiently large to lead to the negative MR, the MR will remain negative upon subsequent field sweeps (Fig. 2). In such a case, only the curve obtained in the first sweep will be different from

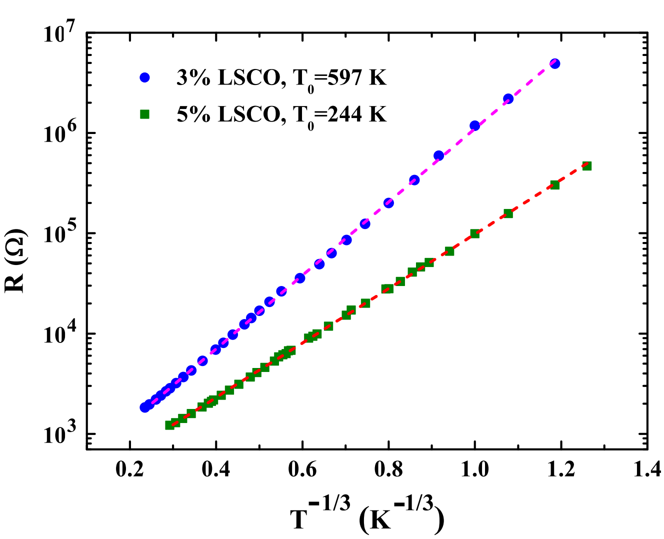

Fig. 1. The temperature dependence of the zero-field cooled in-plane resistance R: below 70 K for 3% LSCO (•) and below 40 K for 5% LSCO (■) samples. Dashed lines are linear fits.

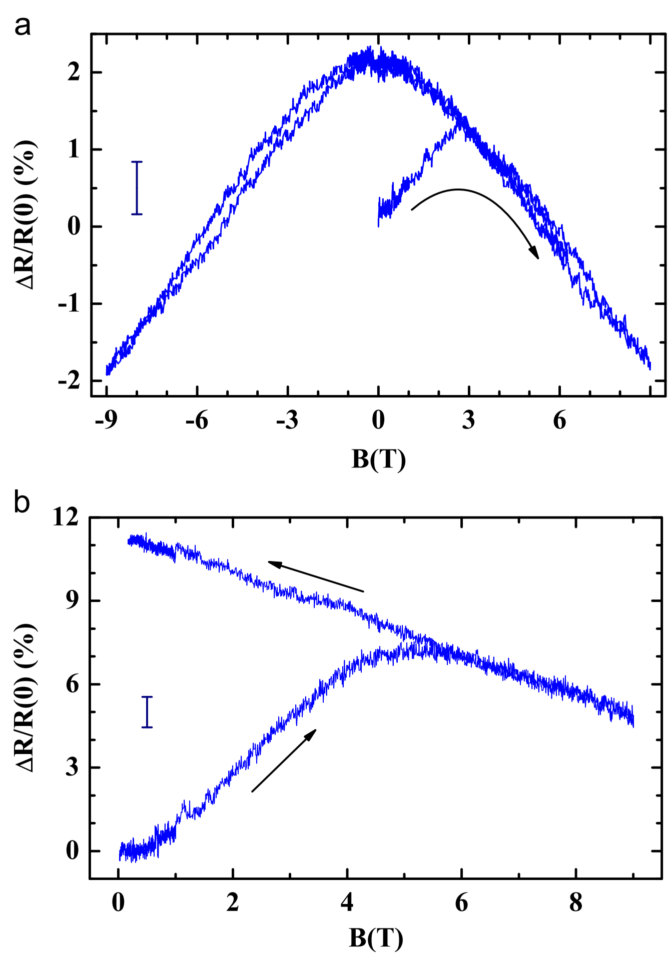

Fig. 2. 3% LSCO. Hysteretic behavior of the positive component of the in-plane MR for $B \parallel c$ at (a) $T=2$ K and (b) $T=0.6$ K. The error bars correspond to the maximum change in the MR due to T fluctuations (6 mK in (a) and 2 mK in (b)). (a) The arrow denotes the direction of the initial sweep from 0 to 9 T. This was followed by sweeps from 9 T to −9 T, then to 6 T, and then back to 0. The first sweep is clearly different from subsequent ones, which are the same within the error. (b) The arrows denote the direction of B sweeps: from 0 to 9 T, then back to 0. In both (a) and (b), the sweep rates were low enough (0.005 T/min for $B<1$ T, 0.02 T/min for $B>1$ T) to avoid the sample heating.