

## Scaling of the Temperature Dependent Hall Effect in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

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A new aspect of the charge dynamics in layered metallic cuprates has been discovered in the study of the in-plane Hall coefficient ( $R_H$ ) in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  over a wide temperature (4–500 K) and composition ( $0 < x < 0.35$ ) range. The strongly temperature dependent  $R_H$ , a hallmark of cuprate superconductors, is found to be the low temperature part of a simple functional form  $f(T/T^*)$  which becomes constant for  $T > T^*$ . This form, followed for  $x \geq 0.15$ , has a characteristic temperature  $T^*$  which is high at the optimal composition ( $x = 0.15$ ) and decreases with increasing Sr concentration, similar to a temperature scale deduced from magnetic susceptibility.

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Layered metallic cuprates are not only superconductors with record high transition temperatures ( $T_c$ ), they are also most remarkable metals in the normal state above  $T_c$  [1–5]. The charge dynamics, for example, is characterized by an in-plane resistivity ( $\rho_{ab}$ ) that grows nearly linearly with temperature from just above  $T_c$  up to the measurement limits of  $\sim 1000$  K [6,7]. The slope of this  $T$ -linear resistivity is quite similar in the various compound families [8]. Equally striking is the strong temperature dependence of the in-plane Hall coefficient ( $R_H$ ), decreasing in magnitude in some cases as fast as  $1/T$  [9]. These and other experimental results have led to the notion that the superconducting cuprates do not behave like Fermi liquids in the normal state [10,11]. For example, the observation of a  $T^2$  dependence of the Hall angle ( $=\sigma_{xx}/\sigma_{xy}$ ) has been interpreted as due to spin-charge separation in a Luttinger liquid [12].

It has been commonly believed that on continued carrier doping beyond the superconducting phase, normal metallic behavior is recovered. However, a recent study of  $\rho_{ab}$  in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (LSCO) for a wide range of carrier concentrations has revealed that unconventional behavior persists to high doping levels [13]. To gain further insight into the charge dynamics of metallic cuprates, particularly in the supposed “normal Fermi liquid” regime for large hole concentrations, we have studied  $R_H$  in LSCO as the hole concentration is varied from  $x = 0.025$  to  $x = 0.34$  in high-quality single-crystal thin films and polycrystalline samples. We find that the much discussed  $T^2$  dependence of the Hall angle is restricted to compositions  $x \leq 0.15$ . Furthermore, we find that for all compositions  $x \geq 0.15$ ,  $R_H$  displays a common functional form  $f(T/T^*)$  which is strongly temperature dependent below  $T^*$  and becomes constant above  $T^*$ . The temperature scale  $T^*$  is high at the optimal Sr composition ( $x = 0.15$ ) and decreases rapidly as the hole concentration increases to the overdoped region ( $x > 0.2$ ). A similar temperature scale can be identified by a broad peak in the static magnetic susceptibility, and this temperature scale follows the same composition dependence as  $T^*$ . Together, these results delineate two different

metallic regions in the phase diagram of this model cuprate system.

The  $c$ -axis-oriented single-crystal thin films with thicknesses of 3500–8000 Å were grown on  $\text{SrTiO}_3$  (100) substrates [14]. The polycrystalline bulk samples were prepared through solid-state reaction processing with repeated regrindings and high temperature annealing [15]. The Hall effect measurements were performed by a dc method in fields of up to 7 T using a six probe geometry. By simultaneously measuring  $\rho_{ab}$  and  $R_H$ , both in duplicate using two pairs of voltage probes each, we were able to verify sample homogeneity and measurement reproducibility. The field independence of  $R_H$  was verified up to 7 T, and most of the measurements were taken at 4 T. The simultaneously measured  $\rho_{ab}$  were found to be in good agreement with previously published results [13]. The measurements above room temperature were taken with a separate probe that provided a flowing oxygen atmosphere to ensure sample integrity.

The evolution of  $R_H$  upon Sr doping is given in Fig. 1. It has previously been observed that the magnitude and temperature dependence of  $R_H$  in polycrystalline samples is dominated by that of the in-plane  $R_H$  measured in single crystals, so both data can be directly compared [2]. We have further confirmed this observation by comparing the data from our single-crystal thin films with the data from our polycrystalline samples at various compositions throughout the range studied. At low doping, the magnitude of  $R_H$  drops rapidly with increasing Sr content, consistent with increasing hole concentration [16]. At the optimal composition,  $R_H$  is strongly temperature dependent to beyond 500 K. This is remarkable in the context of a conventional Boltzmann-type picture, where temperature dependence can be caused by an anisotropic scattering time [ $\tau_k(E)$ ] which changes with temperature. When electron-phonon scattering dominates transport, the scattering at low temperatures is restricted to areas of the Fermi surface which allow small-momentum transfer. As temperatures increase, this kinematic restriction is relaxed, and above a fraction of the Debye temperature (usually  $0.2\theta_D$ – $0.4\theta_D$ ) the scattering becomes isotropic

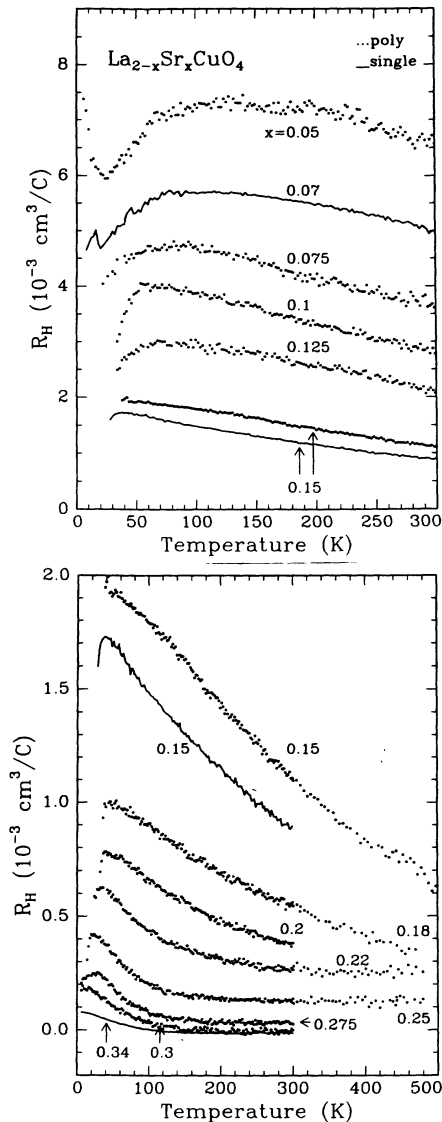


FIG. 1. The temperature dependence of the Hall coefficient for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  with  $0 < x < 0.35$ . Solid lines denote single-crystal data with field  $\mathbf{H} \parallel c$ ; dots denote polycrystalline data.

and temperature independent, leading to a constant  $R_H$ . In the case of LSCO, however, the temperature dependence persists far above any phonon related temperature, such as  $\theta_D$  (thermal  $\theta_D \approx 400$  K), or even the highest phonon frequency [17]. Thus, the electron-phonon scattering is unlikely to be the cause of the observed strongly temperature dependent  $R_H$ .

As the Sr content increases beyond the optimal composition, a systematic evolution can be discerned in  $R_H$ .  $R_H$  is temperature dependent at low temperatures and then approaches a high temperature asymptotic value  $R_H^\infty$ . As doping is increased, this high temperature value decreases and crosses zero near  $x \approx 0.3$  [18]. In addition, the temperature dependence of  $R_H$  evolves with doping by a diminishing characteristic temperature scale. All of the

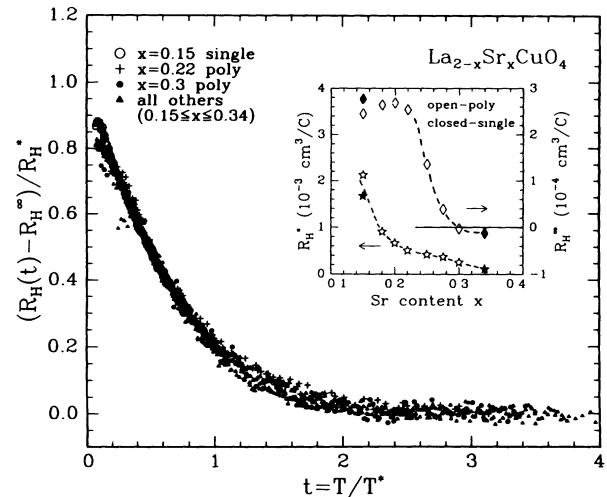


FIG. 2. The Hall coefficient ( $R_H$ ) for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  with  $0 \leq x \leq 0.34$ , plotted rescaled as  $[R_H(t) - R_H^\infty]/R_H^*$  vs  $t$ , where  $t = T/T^*$ .  $R_H^\infty$  is the high temperature limit of  $R_H$ ,  $R_H^*$  rescales the magnitude, and  $T^*$  is the temperature scale. Inset: The parameters  $R_H^\infty$  and  $R_H^*$  vs Sr composition  $x$ . Note the rapid dropoff of  $R_H^*$  above  $x = 0.2$ .

data have roughly the same functional form, expressed by  $R_H^\infty + R_H^* f(T/T^*)$ , where  $f(T/T^*)$  is the purely temperature dependent part of  $R_H$ , which goes to zero at high temperatures.  $R_H^*$  rescales the magnitude of  $f(T/T^*)$  and  $T^*$  rescales the temperature for a given composition. We have defined  $T^*$  to be the intercept between the low temperature behavior of  $R_H$  and the high temperature limiting value of  $R_H$ . Thus  $T^*$  for a given composition marks approximately the crossover from a temperature dependent to a temperature independent  $R_H$ . Although this definition is somewhat arbitrary, the scaling behavior of  $R_H$  allows  $T^*$  to be quantitatively compared between different compositions. [Slightly different definitions of  $T^*$  would yield the same composition dependence observed here. A simple mathematical expression describing  $f(T/T^*)$  could not be found. For example, the expression  $A + B/(T + \theta)$  is not adequate.]

The Hall coefficient rescaled as  $[R_H(T/T^*) - R_H^\infty]/R_H^*$  is shown in Fig. 2 for all compositions  $x \geq 0.15$ . Such a parametrization appears to represent the data quite well. Two characteristic quantities can be derived from Figs. 1 and 2. The first is the high temperature limit  $R_H^\infty$ , shown in the inset to Fig. 2. (The values for  $x \geq 0.2$  are well determined from the data directly, while for  $x < 0.2$  they are significantly affected by the uncertainty inherent in the extrapolation to highest temperatures.)  $R_H^\infty$  has been examined in an exact high temperature expansion of the  $t$ - $J$  model [19]: For the two dimensional square lattice,  $R_H$  is calculated to be positive (despite the electronlike Fermi surface), monotonically decreasing with doping, and to undergo a sign change around  $\frac{1}{3}$  hole filling, which agrees well with the observed change of sign for  $R_H^\infty$  at  $x \approx 0.3$ .

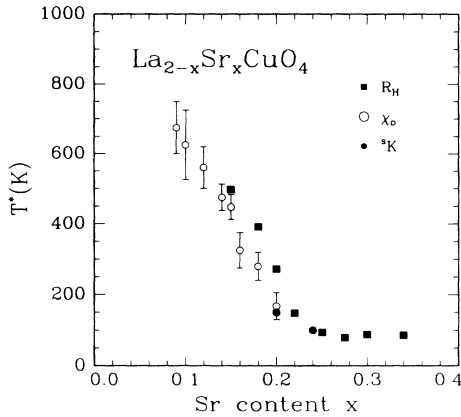


FIG. 3. The characteristic (peak or crossover) temperature  $T^*$  derived from  $R_H$ ,  $\chi$ , and Knight shift vs Sr content  $x$ .  $\chi$  data are from Ref. [20]. Knight shift data are from Ref. [21].

The second quantity of interest is the characteristic temperature  $T_{R_H}^*$ . This temperature scale, which dominates  $R_H$ , varies rapidly with hole concentration and is very high near the optimal concentration ( $\sim 500$  K for  $T_{R_H}^*$  as defined here), but drops rapidly as the (nonsuperconducting) overdoped region is approached. Thus the extended temperature dependence of  $R_H$  is closely associated with the occurrence of superconductivity, just as is the extended  $T$ -linear resistivity. The scaling of  $R_H$  argues against a conventional explanation of the temperature dependence based on dominant electron-phonon scattering, even in the overdoped region. Although for  $x \geq 0.25$   $R_H$  loses its temperature dependence by  $\sim 90$  K (which corresponds to  $\sim 0.2\theta_D$ ), the fact that it has the same functional dependence as for  $x < 0.25$  brings up the questions discussed previously for the optimal composition. The same mechanism dominating  $R_H$  at  $x=0.15$  appears to scale down in temperature with higher doping.

We find that a similar diminishing temperature scale can be found in other measurements as well. It has previously been observed that there is a broad peak in the static susceptibility ( $\chi$ ) for compositions  $x=0.09$  to  $0.2$ , and that the temperature for this peak ( $T_\chi^*$ ) decreases with increased doping [20]. This temperature roughly marks the crossover from  $d\chi/dT > 0$  below  $T_\chi^*$  to  $d\chi/dT < 0$  above  $T_\chi^*$ . Above  $x=0.2$ , it is difficult to discern this maximum in the statically measured  $\chi(T)$  due to a growing low temperature upturn. However, more recent studies of the Knight shift in the overdoped region show a similar peak in temperature ( $T_K^*$ ) that is in very good agreement with the susceptibility data at  $x=0.2$ , and this peak continues to decrease with increased doping [21].

Figure 3 summarizes these various characteristic temperatures and illustrates a central result of this study: The crossover temperature for  $R_H$  ( $T_{R_H}^*$ ) and the peak temperature for  $\chi$  ( $T_\chi^*$ ) and the Knight shift ( $T_K^*$ ) follow essentially the same behavior, and these characteristic temperatures decrease with increased doping. This trend of a rapidly decreasing energy scale appears to occur

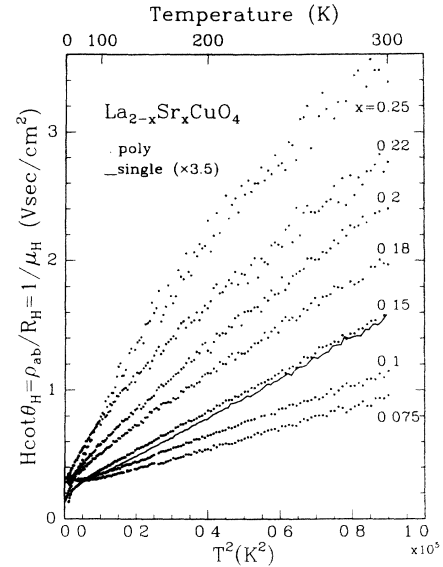


FIG. 4.  $H \cot \theta_H$  as a function of  $T^2$  for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  with  $0.075 \leq x \leq 0.25$ . Solid lines denote single-crystal data (multiplied by a factor of 3.5); dots denote polycrystalline data.

throughout the Brillouin zone, since  $\chi$  (and the Knight shift) probes near  $\mathbf{q}=0$  while  $R_H$  examined in a Boltzmann picture is a complex average over  $\mathbf{q}$ . It is intriguing that this observed characteristic temperature decreases rapidly as bulk superconductivity is lost [15]. Recent studies show a rapid decrease of the resistivity anisotropy ( $\rho_c/\rho_{ab}$ ) in the overdoped region, which has been interpreted as the evolution from a two dimensional metal to an anisotropic three dimensional metal [22,23]. The sharp decrease in  $T^*$ , the loss of bulk superconductivity, and the rapidly diminishing resistivity anisotropy all occur in the same range of Sr composition.

In addition to the rescaling of  $R_H$  discussed above, we have also analyzed the temperature dependence of the Hall angle ( $\theta_H$ ), a quantity of recent interest due to the observation of a simple temperature dependence of the form

$$\cot \theta_H = \sigma_{xx} / \sigma_{xy} = A + BT^2 \quad (1)$$

for LSCO and YBCO [12,24]. It has been suggested that  $\cot \theta_H \propto 1/\tau_H$ , where  $\tau_H$  is a new transverse relaxation rate that is distinctly different from  $\tau_{tr}$  determining the longitudinal transport  $\rho_{ab}$  [25]. Two scattering times arise in the context of the spin-charge separated Luttinger liquid state. A plot of  $H \cot \theta_H = \rho_{ab}/R_H$  (also equal to the inverse Hall mobility,  $\mu_H$ ) versus  $T^2$  is given in Fig. 4. For  $x \leq 0.15$ , the data follow a  $T^2$  fit for  $T > 100$  K, with the systematic trend that the coefficient of the  $T^2$  term increases with doping. The deviation from  $T^2$  at low temperatures for low Sr compositions may be attributed to the onset of weak localization. The data for  $x=0.15$  follow quite closely a  $T^2$  form to at least 500 K (not shown). At compositions with  $x > 0.15$ , however,

the data cannot be described by  $A + BT^2$  over any appreciable temperature range between 4 and 500 K, even for compositions that exhibit bulk superconductivity.

There is currently no theoretical result exploring how  $R_H$  or  $\rho_{ab}$  should evolve in the overdoped regime within the framework of the proposed Luttinger liquid. In the limit of high doping it is expected that a conventional Fermi liquid with a single  $\tau$  would be recovered, but questions of how  $\tau_H$  and  $\tau_{tr}$  (spin and charge degrees of freedom) recombine and how quickly this should occur have not been addressed. Our data give some empirical results related to this issue. It is unclear how to interpret the deviation of  $\cot\theta_H$  from  $T^2$  for  $x \geq 0.18$  within the spin-charge separation picture. Furthermore, the zero crossing of  $R_H$  near  $x = 0.3$ , which leads to a divergence in  $H \cot\theta_H$ , makes any interpretation of  $\theta_H$  difficult. The most distinct observation is the simple scaling of  $R_H$  in the overdoped region. A new  $\tau_H$  was invoked because  $\theta_H$  appears to obey a simple Matthiessen rule with regard to impurity scattering, and from this it was inferred that  $\theta_H$  is an intrinsic quantity, while  $R_H$  is not because it mixes both scattering times,  $R_H \propto \tau_{tr}/\tau_H$ . However, our discovery of the scaling of  $R_H$  indicates that in the overdoped region  $R_H$  itself is a quantity of primary interest. It remains unclear whether it is appropriate to interpret all aspects of the charge dynamics in terms of a temperature, frequency, and doping dependent scattering rate without considering an evolution in the quasiparticle states themselves.

In summary, the present extensive study of the in-plane Hall effect to high temperature for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  has expanded and clarified the understanding of the highly unusual charge dynamics in this layered metallic cuprate. The strongly temperature dependent Hall coefficient, a hallmark of the high  $T_c$  cuprates, was found to evolve systematically with increasing hole concentration in the optimal and overdoped regime ( $x \geq 0.15$ ). Above a characteristic temperature  $T^*$ ,  $R_H$  tends to be constant ( $R_H^\infty$ ), and it increases rapidly below  $T^*$ . This characteristic temperature is high for the optimal composition ( $x = 0.15$ ), decreases rapidly with further doping, and saturates at a low value above  $x \approx 0.22$ . A next step will be to elucidate quantitatively the relationship between the electronic and magnetic degrees of freedom ( $T^*$  is observed in both) over such a wide hole concentration range.

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*Note added.*—After submission of this manuscript, the authors have learned of a similar study of the Hall coefficient in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  over an extended temperature range (100–1000 K) [26]. Application of our analysis results in the same values for  $T^*$  within experimental error.

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