

EXPERIMENTAL EVIDENCE FOR MULTI-BAND CONDUCTION IN HIGHLY-DOPED La-Sr-Cu-O SUPERCONDUCTORS

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We have measured the Hall coefficient in highly-doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ superconductors ($x=0.2$ and 0.25), finding a large decrease with temperature. A good fit to the data is obtained using the two-band model involving both holes and electrons (as suggested by our earlier thermopower measurements), with the carrier-phonon interaction being relatively stronger for the holes.

Much insight has been gained over the past few months into the physical properties of the recently discovered high T_c superconductors based on the oxygen-deficient perovskites. To help elucidate the focal point, namely the nature of the coupling mechanism responsible for the unprecedentedly high T_c observed in this class of materials, a wide range of experimental techniques has been brought to bear on this problem. Pivotal among these studies are investigations of the normal state behavior, which provide fundamental input parameters for various theories of superconductivity. Perhaps potentially the most profitable among these are measurements of the Hall coefficient, which yield not only an estimate of the carrier density and identify the polarity of the dominant carriers, but also point out, via the temperature dependence, any unusual carrier scattering features and multiplicity of the carrier spectrum. While in principle the Hall effect measurements are very simple, the extremely small mobilities of a few $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$ typical of these ceramics result in very small Hall voltages that have to be carefully dis-

cerned from a large and strongly temperature dependent resistive voltage. This complicates the measurements and the available data on the temperature dependence are, so far, by no means plentiful.

Panson et al. [1] measured a Hall coefficient of magnitude $(5 \pm 2) \times 10^{-9} \text{ m}^3 \text{ C}^{-1}$ on a sample of La-Sr-Cu-O ($x=0.2$) at 48 K in a field of 5 T. Most surprisingly, these authors report the Hall constant as negative, contrary to an intuitive picture of hole-dominated transport and subsequent Hall effect investigations. Ong et al. [2] reported a strong compositional dependence of the Hall effect measured at 77 K on a series of La-Sr-Cu-O compounds spanning the Sr content range between $x=0.05$ and 0.25 . They noted a precipitous fall in the Hall constant setting in at $x=0.15$, and stated that above $x=0.18$ the signal became undetectably small in spite of a 15 T transverse field used in these studies. Hundley et al. [3] made investigations on a sample of La-Sr-Cu-O ($x=0.15$) which indicate a linearly decreasing Hall constant with increasing temperature. In addition, they noted an interesting sharp "spike" on the data just as the sample goes superconducting, which they

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scribe to an enhanced magnetic field in the normal regions of the sample as the flux is excluded from the superconducting grains. A modest temperature dependence of the Hall coefficient in La-Sr-Cu-O for $x=0.15$ and $x=0.185$ was also found by Penney et al. [4].

In the course of our recent studies on the electronic and thermal transport properties of La-Sr-Cu-O superconductors [5] we have observed a sign change (from positive to negative) in the thermopower of the most heavily Sr-doped compounds with increasing temperature and we have pointed out the likelihood of a multicarrier nature of the carrier spectrum in these materials. To substantiate this point we have measured the temperature dependence of the Hall coefficient on these samples and we describe the results in this Letter.

dc Hall measurements were made on samples of La-Sr-Cu-O with $x=0.2$ and $x=0.25$ cut from the same disks as used for the thermopower studies. The samples were thinned down to about 0.25 mm thickness and copper leads were attached with the aid of silver epoxy. Samples were coated with Apiezon grease and fastened with a dental floss to a cigarette paper-insulated copper cold tip of a cryostat. The five-probe (resistive voltage cancellation) as well as four-probe technique with a field reversal were employed and gave comparable results. The data were taken with a current density of 0.5 A cm^{-2} in a field of 7 T. To make sure that the temperature of the samples is the same after the field is reversed from +7 to -7 T (about 10 minutes procedure) a small, flat Pt thermometer was attached to each sample using Apiezon grease. While Pt has a substantial magnetoresistance, at a fixed field its resistance is immune to the field reversal. The base temperature was monitored by a calibrated glassy-carbon sensor.

Our results are shown in fig. 1, together with the earlier data [3] for a $x=0.15$ sample. It is clear that the trend of the two data sets is consistent. What our new data show is that besides reducing in size as x increases, the Hall coefficient R_H shows a much stronger relative temperature dependence. For the $x=0.25$ sample, the decrease in R_H between 40 and 200 K is nearly 70%. These measurements provide evidence that the widely used one-band model is not applicable for the interpretation of the Hall effect at least in the highly doped La-Sr-Cu-O supercon-

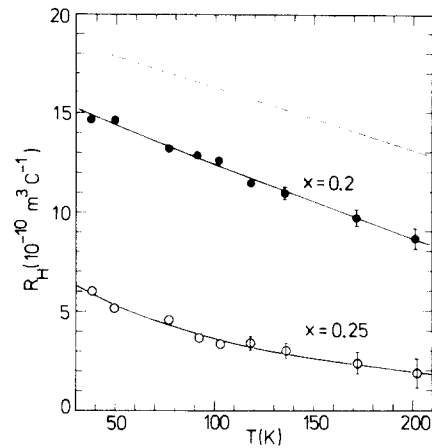


Fig. 1. Our measurements (circles) of the Hall coefficient in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ with $x=0.2$ and $x=0.25$, with the results of Hundley et al. [3] for $x=0.15$ for comparison (dotted line). The full lines are calculations for the two-band model with parameters $\rho_h(100) = 540 \mu\Omega \text{ cm}$, $\rho_h(200) = 1030 \mu\Omega \text{ cm}$, $\rho_c(100) = 4090 \mu\Omega \text{ cm}$, $\rho_c(200) = 4280 \mu\Omega \text{ cm}$, $n_h = 3.7 \times 10^{21} \text{ cm}^{-3}$, and $n_e = 1.0 \times 10^{21} \text{ cm}^{-3}$ for the $x=0.2$ sample, and $\rho_h(100) = 720 \mu\Omega \text{ cm}$, $\rho_h(200) = 1280 \mu\Omega \text{ cm}$, $\rho_c(100) = 1660 \mu\Omega \text{ cm}$, $\rho_c(200) = 2360 \mu\Omega \text{ cm}$, $n_h = 5.2 \times 10^{21} \text{ cm}^{-3}$, and $n_e = 2.5 \times 10^{21} \text{ cm}^{-3}$ for the $x=0.25$ sample.

ductors, and that it is not possible in these cases to obtain reliable values for the density of carriers using this model.

In strong contrast to the case of pure La_2CuO_4 [6] and lightly Sr-doped compounds [7], the transport properties for La-Sr-Cu-O with x greater than 0.15 are consistent with those expected for ordinary metallic conduction but with a small density of carriers [5]. It is, of course, likely that the electrons in these superconductors are moderately strongly correlated [8,9], but the correlations do not appear strong enough to change the linear metallic resistivity dependence, for example. It therefore seems worthwhile to attempt to account for the Hall coefficient behaviour in terms of the usual metallic two-band expression [10]

$$R_H = \left(\frac{\rho}{\rho_h}\right)^2 \frac{1}{n_h e} - \left(\frac{\rho}{\rho_c}\right)^2 \frac{1}{n_c e}. \quad (1)$$

Here n_h and n_e are the carrier densities (assumed independent of temperature T) for the hole and electron bands respectively, e is the magnitude of electronic charge, ρ_h and ρ_c are the corresponding re-

sistivities for the two bands, and ρ is the total resistivity

$$\rho = \rho_h \rho_e / (\rho_h + \rho_e) . \quad (2)$$

The band resistivities ρ_h and ρ_e are taken as linear in the temperature range of our Hall measurements (approximately 40 to 200 K); we write them in the form

$$\rho_i = (2 - T/100)\rho_i(100) - (1 - T/100)\rho_i(200) \quad (3)$$

$(i=h, e) ,$

since it is convenient to characterize the resistivities ρ_i by their values $\rho_i(100)$ and $\rho_i(200)$ at temperatures 100 and 200 K respectively. The observed values of the total resistivity ρ at these temperatures provide two constraints for the four parameters $\rho_h(100)$ and $\rho_h(200)$, but since both the observed and calculated resistivity (for the parameter ranges of interest) are approximately linear, two degrees of freedom remain.

As shown by the examples in fig. 1, the two-band model can give an excellent account of the data for reasonable parameter values. Even the sign of the curvature apparent in the $x=0.25$ data is consistent with the two-band model prediction. Since the values of the parameters are not uniquely determined by the data, the precise values for n_h and n_e are not significant. However, the essential features required for the two-band model to fit our experimental data are as follows.

Because R_H is positive, holes are the dominant carriers, with a density of a few times 10^{21} cm^{-3} . The temperature dependence of R_H in our calculation arises because the contribution from the electron band is weighted more strongly as temperature increases, i.e. because ρ_h for the hole band increases with T relatively faster than ρ_e for the electron band; for example, we find $\rho_h(200)/\rho_h(100) = 1.79$ compared to $\rho_e(200)/\rho_e(100) = 1.42$ for the parameters pertaining to the $x=0.25$ curve in fig. 1, and 1.89 and 1.05 respectively for the parameters for the $x=0.2$ sample. Such behavior arises if the coupling between the holes and phonons is much stronger than that between electrons and phonons, leading to a faster increase with T of ρ_h than ρ_e relative to the corresponding "residual" resistivities (a smaller structural scattering of the holes could also contribute to

this effect). A strong interaction of holes and phonons is consistent with our earlier conclusion [5] from the size of the temperature dependence of resistivity. The effect of the electrons is clearly more significant for $x=0.25$ than for the lower concentrations, indicating that the density of electrons increases faster with Sr concentration x than that of holes.

For the parameter values listed in the caption to fig. 1, and for ρ_h and ρ_e assumed linear in the temperature range of interest, the total resistivity ρ shows a very slight downward curvature below a linear law at lower temperatures. Such behavior, however, would likely be masked in practice by the upward curvature of the electron-phonon contribution that typically occurs below about 50 K. Such an upward curvature does appear to be observed [5] for the $x=0.25$ sample with the lower T_c value, but superconducting precursor effects soon produce a large decrease in ρ as T decreases.

In conclusion, we have demonstrated strong evidence for multi-band conduction in highly doped La-Sr-Cu-O. The fact that our present and previous data [5] are explicable in terms of usual metallic conduction with a strong hole-phonon interaction without the need for exotic mechanisms suggests that this hole-phonon coupling could at least play a role in the origin of the high T_c superconductivity.

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