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Temperature dependence of the Hall effect in single-layer and bilayer $Bi_2Sr_2Ca_{n-1}Cu_nO_y$ thin films at various oxygen contents

Z. Konstantinović, Z. Z. Li, and H. Raffy
Laboratoire de Physique des Solides, Bâtiment 510, Université Paris-Sud, 91405 Orsay Cédex, France
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The Hall coefficient $R_H(T)$ is studied in epitaxial *c*-axis-oriented single-layer $Bi_2Sr_{1.6}La_{0.4}CuO_y$ and bilayer $Bi_2Sr_2CaCu_2O_y$ thin films at various oxygen concentrations going from overdoped to underdoped. In both phases, the Hall angle obeys the law $B+CT^\alpha$ $(1.65\leqslant\alpha\leqslant 2)$, with α increasing with decreasing oxygen content, yielding a T^2 dependence only in the underdoped region. The temperature T_0 below which an upward deviation from this law occurs is compared to the pseudogap temperature T^* deduced from resistivity $\rho(T)$. The manifestation of the pseudogap opening in the Hall effect is discussed.

The evolution of the electronic properties of cuprates with hole doping and the pseudogap phenomena observed in the underdoped region[1] are intensively studied to understand their normal-state properties, generally considered to be closely related to the superconducting mechanism. The temperature dependence of the Hall coefficient R_H (Ref. 2) is one of the unusual properties of the normal state of high-T_c superconductors. The Hall angle, defined by $\cot\theta_H=\rho/R_HH$ where H is the magnetic field and ρ the resistivity, was shown to present a simpler temperature behavior than R_H.[3] However, subsequent detailed experimental studies of $\cot\theta_H(T)$ at various doping revealed a more complicated behavior[4,5] than the previously reported Fermi-liquid-like T^2 dependence.[6] It was also shown that, above a characteristic temperature T_0, the Hall angle obeys a power law T^α with α decreasing from 2 with increasing carrier concentration.[7,8] Moreover T_0 is considered by some authors as the characteristic temperature below which the pseudogap affects the Hall angle,[9] in contrast with reports noting that the pseudogap affects selectively the in-plane resistivity and leaves unchanged the Hall angle.[10]

Here, we present a systematic study of the temperature dependence of the Hall coefficient and of the Hall angle from maximally overdoped to strongly underdoped nonsuperconducting states measured on epitaxial *c*-axis oriented single-layer $Bi_2Sr_{1.6}La_{0.4}CuO_y$ and bilayer $Bi_2Sr_2CaCu_2O_y$ thin films. In both cases, the temperature region $(T\geqslant T_0)$ where $\cot\theta_H$ obeys a law of the form $B+CT^\alpha$ is determined and its low-temperature limit T_0 is compared with the pseudogap temperature T^* determined previously from the in-plane resistivity.[11]

The samples are *c*-axis-oriented epitaxial $Bi_2Sr_{1.6}La_{0.4}CuO_y$ [Bi(La-2201)] and $Bi_2Sr_2CaCu_2O_y$ (Bi-2212) thin films, grown by rf magnetron sputtering,[12] with thickness ranging from 1000 to 2000 Å. The partial substitution of Sr by La in $Bi_2Sr_{2-x}La_xCuO_y$ increases the maximal critical temperature, T_{cmax}, from 18 K for $Bi_2Sr_2CuO_y$ $(x=0)$ to 30 K for $x=0.4$ and enlarges the underdoped region. Besides, the lower T_{cmax} value of Bi(La)-2201 compared to that of Bi-2212 allows us to study the normal state in a wider temperature range. The films were patterned into a stripline equipped with six gold sputtered contact pads. The in-plane resistivity was measured using a dc standard four-probe method at different oxygen doping levels. The Hall coefficient R_H was measured using a classical field inversion technique under a magnetic field of 1 T parallel to the *c* axis of the film. We checked at room temperature, up to 20 T, that R_H is independent of the applied magnetic field. The oxygen content of a given sample was changed by repeated annealing treatments in a controlled atmosphere going from a maximally overdoped state [$T_c(R=0)=13$ K for Bi(La)-2201 and $T_c(R=0)=50$ K for Bi-2212] to strongly underdoped nonsuperconducting states $(T_c=0)$.[11] We verified that the transport properties of the states obtained in this way are fully reproducible. The number of holes per Cu, *p*, is evaluated from the phenomenological law $T_c=T_{cmax}[1-82.6(p-0.16)^2]$.[13]

The resistivity data for such Bi-2212 and Bi(La)-2201 thin films, spanning a wide range of doping levels, are published elsewhere.[11] Briefly, the overdoped states can be described by a phenomenological law of the form $\rho=\rho_0+AT^m$, with the exponent m decreasing from its maximal value $m_{max}=1.3$ to 1 corresponding to the optimal state $(T_c=T_{cmax})$. In the underdoped region, a pseudogap opens in the electron excitation spectra, which causes a downward deviation of the resistivity from its high-T linear behavior. In the strongly underdoped region localization effects occur and compete with pseudogap effect.

The behavior of the Hall coefficient R_H vs T from a maximally overdoped state up to a strongly underdoped state $(T_c=0)$ is shown in Fig. 1(a) for Bi(La)-2201 and Fig. 1(b) for Bi-2212 thin films. For both phases, the curves $R_H(T)$ show a maximum at $T=T_{max}$ for all doping levels. In the overdoped region, the values of the characteristic temperature T_{max} are smaller for Bi(La)-2201 than for Bi-2212 due to very different T_c values and they both increase with reducing carrier concentration. In the underdoped region, T_{max} values of both phases are very similar and almost T independent with $T_{max}\sim 135\pm 10$ K.

In order to compare the Hall effect for both phases, we show in Fig. 2(a), for a variety of samples, the doping dependence of the quantity R_HeN/V, inversely proportional to the Hall number per Cu, where N is the number of Cu atoms per unit cell of volume V and e the electronic charge. The

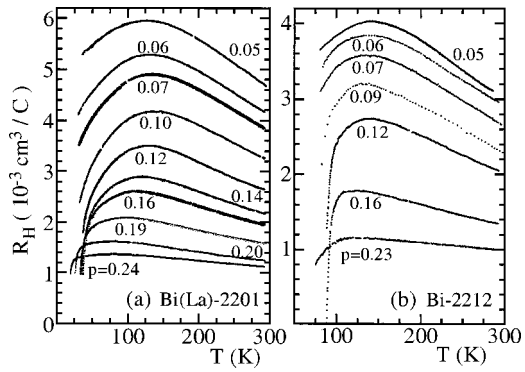

FIG. 1. Temperature dependence of R_H for (a) Bi(La)-2201 and (b) Bi-2212 thin films at different doping levels from maximally overdoped to strongly underdoped states ($T_c=0$). The results for the Bi(La)-2201 phase are obtained from one film, while the results for the Bi-2212 phase are obtained from three different films: one optimally doped ($p=0.16$), one underdoped ($p=0.12$), and one film in various doping states ($p=0.23$, 0.09, 0.07, 0.06, and 0.05).

systematic increase of R_H with decreasing oxygen contents observed for both phases indicates a decrease of carrier numbers of the order of 4 at room temperature between the two extreme states. It is in good agreement with the decrease of p from 0.22 to 0.05. This result indicates that the Hall number per Cu is proportional to the calculated number of holes per Cu, p, the factor of proportionality being of the order of 4. Note also that the absolute magnitudes of R_HeN/V for both phases agree reasonably well with the values found in $Bi_2Sr_{1.61}La_{0.39}CuO_{6+\delta}$ single crystals as well as in Y-Ba-Cu-O and La-Sr-Cu-O compounds, as reported in Ref. 14. The dispersion of ~20% between the values of the two phases (Fig. 2) comes mainly from the incertitude in geometrical parameters as thickness and width of the samples. Moreover, the temperature dependence of R_HeN/V, shown for optimally doped states [Fig. 2(b)] for both phases, presents a very similar behavior for $T>120$ K, while the difference seen at low temperature comes from the higher T_c value of the Bi-2212 phase as indicated above.

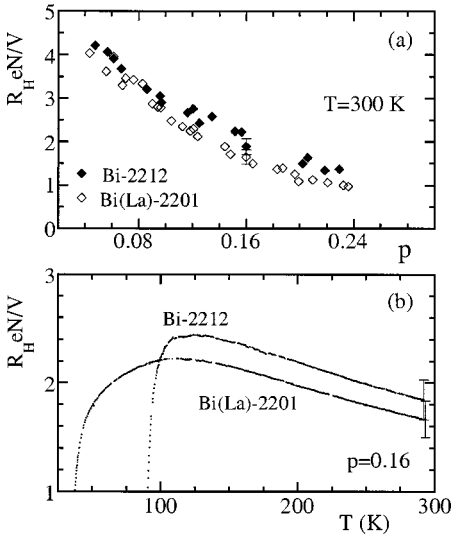

FIG. 2. The values of R_HeN/V as a function of (a) doping at room temperature and (b) temperature for optimally doped states for Bi(La)-2201 and Bi-2212 thin films.

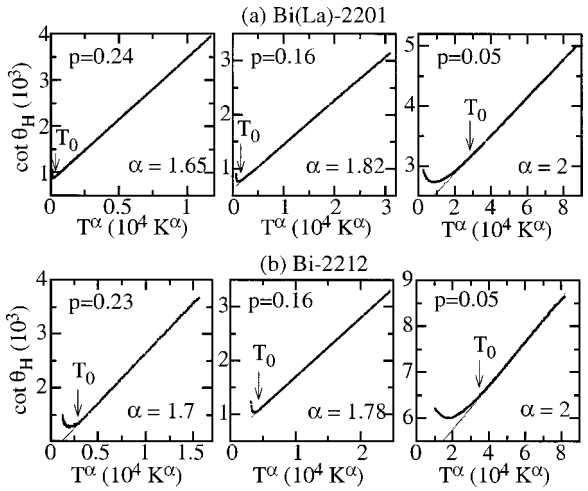

FIG. 3. The evolution of cot θ_H vs T^α for some characteristic doping states of (a) Bi(La)-2201 and (b) Bi-2212 thin films. The continuous lines show the curve fit to a phenomenological law $B+CT^\alpha$ with deviation from this behavior at T_0 (see arrows).

Figure 3 shows the evolution of cot $\theta_H(T)$ for three typical doping levels for (a) Bi-(La)-2201 and (b) Bi-2212 films. It appears that the previously found T^2 law can describe only the strongly underdoped states ($p<0.10$) while, for larger p values, cot θ_H obeys a phenomenological T^α law from 300 K down to a characteristic temperature T_0 (marked by arrows) with α less than 2.

Importantly for both phases, the exponent α exhibits the same doping dependence. It decreases linearly from the value 2 to the value 1.65–1.7 for the most overdoped state [Fig. 4(a)]. The large error bars for strongly underdoped states are shown in the same figure to indicate a possible description with an exponent α larger than 2, as it was reported previously in the case of the $Bi_2Sr_{1.26}La_{0.74}CuO_{6+\delta}$ single crystal ($\alpha=2.05$) (Ref. 15) and the $YBa_2Cu_3O_{6.63}$ compound ($\alpha=2.15$).[16] The doping dependence of T_0 is shown in Fig. 4(b). In both phases, the characteristic temperature T_0 increases with decreasing doping in a similar way in the underdoped region where it becomes comparable for Bi-2212 and Bi(La)-2201. In the overdoped region, T_0 is smaller in Bi(La)-2201 than in Bi-2212, which is related to the difference in their critical temperatures. The decrease of T_0 is obstructed in the overdoped region by the vicinity of the superconducting state where T_0 becomes of the same

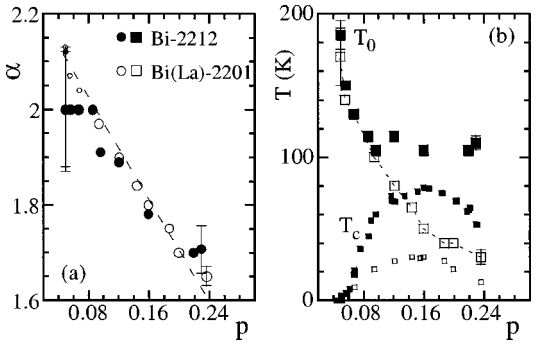

FIG. 4. The doping dependence for both phases of (a) the exponent α and of (b) the characteristic temperature T_0 (the dashed line is a guide to the eye).

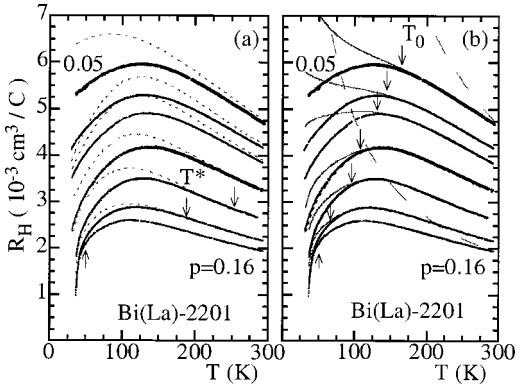

FIG. 5. Hypothetical behaviors of the Hall coefficient given by (a) $R_H=(\rho_0+AT)/H\cot\theta_H$ (dotted lines) and (b) $R_H=\rho(T)/H(A+BT^{\alpha})$ (continuous lines) and $R_H\sim T/T^{\alpha}$ for ''ideal samples'' (dashed lines) for the optimally doped and underdoped states shown in Fig. 1(a).

order of magnitude as T_c. The fact that we have $T_{c2201}<T_{c2212}$ allows one to follow the nearly linear decrease of $T_0(p)$ down to lower temperatures. In addition, it has to be remarked that T_0 is smaller than T^*, determined by resistivity measurements (see below Fig. 5), and we underline that there is no change in the Hall angle at T^* (see Fig. 3).

The above results indicate that the T^2 law of the Hall scattering rate is systematically violated in overdoped and optimally doped regions. Diffusion by spin excitations (spinons) with a Fermi-liquid-like T^2 relaxation rate, predicted in the model of Anderson,[3] can explain the experimental data only in the underdoped region. To take into account the decrease of exponent α one must introduce a scattering source, which changes with doping level. On the other hand, our results are not incompatible with the scenarios which consider a strongly anisotropic relaxation time as in the hot spot model[17] allowing more complicated temperature dependence, although until now to our knowledge there are no theories which explain the monotonic decrease of exponent α with increasing doping. Note that, for practical purposes, the value of α [with $\alpha\sim 2.25p-2.7p$, given by the dashed line in Fig. 4(a)] can be used to characterize the doping level of Bi-2212 and Bi(La)-2201 phases.

As to the characteristic temperature T_0, it was also considered in a recent study as the temperature where the pseudogap appears in the Hall effect and it was related to the temperature where $R_H(T)$ is maximum.[9] In order to check this idea we have examined the hypothetical behavior of the Hall coefficient R_H for Bi(La)-2201 in the absence of the pseudogap effect observed in the temperature dependence of resistivity.[11] In Fig. 5(a) the dotted line represents the variation of $R_H=(\rho_0+AT)/H\cot\theta_H$, where ρ_0+AT describes the high-T linear behavior of $\rho(T)$ reported in a previous article.[11] The deviation between this line and our data occurs at T^*. It can be seen that the maximum is still present in this hypothetical behavior. The characteristic temperature $T_{\max}$ is shifted to lower temperatures, but is not necessarily connected to the characteristic temperature T_0 or to the pseudogap effect. Moreover, we can conclude that the pseudogap opening at T^* reduces R_H below this temperature. Note that the opposite effect was reported in previous studies on Y-Ba-Cu-O compounds where a low-T upward deviation from the $1/T$ dependence of R_H is associated with pseudogap opening.[5] Here there is no clear $1/T$ dependence in $R_H(T)$ in the high-T region due to the presence of constant terms in $\rho(T)$ and in $\cot\theta_H$, the latter seen in Fig. 3. A recent comparison between the different cuprate families reveals very similar temperature dependence of R_HeN/V except for Y-Ba-Cu-O samples.[14] This fact can indicate that the different behavior of $R_H(T)$ for Y-Ba-Cu-O and in particular its change of slope is more possibly related to the properties of the CuO chains which are only present in this compound.

In addition to discuss further the meaning of the characteristic temperature T_0, we consider the hypothetical expression of R_H in the case where $\cot\theta_H$ would continue to change as T^{α} down to T_c. In Fig. 5(b), the continuous lines represent the variation of $R_H=\rho(T)/(B+CT^{\alpha})$, where we take now the experimental behavior of the resistivity. A good agreement with the data is obtained above T_0 with a maximum of R_H present for the states near optimum. As noted above, there is no clear correlation between the two temperatures T_0 and $T_{\max}$. We emphasize also that if we consider just a law for an ''ideal'' sample without residual resistivity and residual Hall angle the experimental results cannot be described at all (see dashed lines in the same figure). The same kind of analysis and the above-discussion are also valid for Bi-2212 phase.

Finally, this characteristic temperature T_0 was associated previously with the lower characteristic temperature for the pseudogap and with the characteristic temperatures deduced from NMR relaxation rate and angle-resolved photoemission spectroscopy measurements.[9,15] Recent comparisons between the different experiments on the Bi-2212 phase have revealed that T^* values determined from in-plane resistivity measurements agree well with those deduced from ARPES and NMR relaxation rates[11,18] and as T_0 is considerably lower than T^* it cannot be identified with the latter. Besides, we note that T_0 and the temperature of the inflection point in $\rho(T)$ ($T_1\sim 0.5T^*$) in the underdoped region show similar doping dependence (although between 20 and 30% smaller).[11]

In conclusion, we have analyzed the evolution of the temperature dependence of Hall coefficients and Hall angles with doping in the normal state of single-layer and bilayer $Bi_2Sr_2Ca_{n-1}Cu_nO_y$ thin films ($n=1,2$). The behavior of the Hall effect is very similar in these two phases with only a difference in the overdoped region coming from their different critical temperatures. While the Hall coefficient shows rather complicated behavior vs T, the cotangent of the Hall angle can be described by a simple phenomenological expression $B+CT^{\alpha}$. The exponent α equal to 2 in the strongly underdoped region decreases with increasing doping to the value 1.65 in the overdoped region. We have established the temperature and doping range where a T^2 behavior is observed. Moreover, no evidence of a pseudogap opening is directly seen in the temperature dependence of the Hall angle at the temperature T^*, determined from resistivity measurements. The signature of pseudogap behavior in $\theta_H(T)$ proposed at T_0 (Ref. 9) is not obvious. It is shown that T_0 is not correlated with $T_{\max}$, determined from the maximum of $R_H(T)$. Finally, these systematic results over a wide region of doping will be useful to test the various theories attempting to give a complete picture of the normal state properties.

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