Hall effect in underdoped $\text{GdBa}_2\text{Cu}_3\text{O}_{7−\delta}$ thin films: evidence for a crossover line in the pseudogap regime.

D. Matthey, S. Gariglio, B. Giovannini, and J.-M. Triscone
Condensed Matter Physics Department, University of Geneva, Switzerland.
(October 28, 2018)

We report on measurements of the resistivity and Hall coefficient in underdoped $\text{GdBa}_2\text{Cu}_3\text{O}_{7−\delta}$ epitaxial thin films grown by off-axis magnetron sputtering. The films have been lithographically patterned allowing precise measurements of the temperature dependencies of the inverse Hall constant $R_H^{-1}$ and of the Hall angle $\theta_H$. We find that $R_H^{-1}$ is linear in temperature between 300 K and the pseudogap temperature $T^*$, whereas $\cot(\theta_H)$ displays a perfect $T^2$ temperature dependence between typically 300 and 100 K. We observe for all the samples that the temperature at which the temperature dependence of $\cot(\theta_H)$ deviates from the $T^2$ behavior is correlated to the temperature at which $R_H$ displays a peak. This characteristic temperature, found to lie between $T_c$ and $T^*$, does not depend markedly on the doping level and defines a new crossover line in the temperature versus doping phase diagram. We tentatively relate these findings to recent high frequency conductivity and Nernst effect experimental results, and we briefly discuss the possible consequences for competing theories for the pseudogap state of the cuprates.

I. INTRODUCTION

Among the anomalous normal state transport properties observed in high temperature superconductors, the temperature dependence of the Hall constant is probably one of the most striking: it displays a behavior which is totally different from the one observed in simple metals. Indeed, it is generally observed, in particular in $\text{REBa}_2\text{Cu}_3\text{O}_{7−\delta}$ (RE rare earth) compounds, that the temperature dependence of the Hall coefficient, $R_H(T)$, goes as $1/T$ over a wide temperature range and presents an ill-understood peak above $T_c$, whereas the cotangent of the Hall angle $\cot(\theta_H) = \rho_{xx}/\rho_{yy}$ where $\rho_{xx}$ and $\rho_{yy}$ are respectively the longitudinal and Hall resistivity) varies as $T^2$ over a wide temperature range. One way to analyze the temperature dependence of the Hall effect is to consider two different relaxation rates. Experimental reports have shown that the cotangent of the Hall angle obeys a $T^2$ law even if $R_H^{-1}(T)$ or $\rho_{xx}(T)$ are strongly nonlinear suggest that the cotangent of the Hall angle is a function of a single relaxation rate, $\tau_H^{-1}$, whereas the resistivity $\rho_{xx}$ would be related to another relaxation rate $\tau_{tr}^{-1}$. Other scenarios based on a temperature dependent carrier concentration or on a scattering time that depends on the Fermi surface location have been proposed. When considering two relaxation rates, the Hall constant $R_H$ depends on both $\tau_H$ and $\tau_{tr}$: $R_H^{-1} = \cot(\theta_H) \cdot H/\rho_{xx} \sim \tau_H^{-1}/\tau_{tr}^{-1}$. The $T^2$ temperature dependence of the cotangent of the Hall angle, $\cot(\theta_H) = AT^2+C$, has been theoretically predicted in, and observed experimentally in many systems. In the approach proposed by Anderson, the two scattering rates reflect spin and charge separation with the Hall angle being directly related to the spinon relaxation rate. In this view, magnetic impurities bring another scattering channel for the spinons and thus directly affect the behavior of the Hall angle as first observed experimentally by T. R. Chien, Z. Z. Wang and N. P. Ong.

Recent experimental results and theoretical ideas on the nature of the pseudogap $\delta$ and its influence on Hall measurements, the possible presence of charge inhomogeneities, or the presence of vortex like excitations in the pseudogap phase, have changed our view of the normal state. In this paper, we revisit the question of the Hall effect by measuring the Hall constant and the Hall angle in the pseudogap phase of epitaxial underdoped $\text{GdBa}_2\text{Cu}_3\text{O}_{7−\delta}$ films. The results are presented and discussed in the context of the recent findings mentioned above.

II. SAMPLES AND EXPERIMENT

A series of $\text{GdBa}_2\text{Cu}_3\text{O}_{7−\delta}$ thin films were grown by off-axis magnetron sputtering on (100) $\text{SrTiO}_3$ substrates. The temperature of the substrates was typically 700 °C, and the $\text{Ar} + \text{O}_2$ sputtering pressure 0.15 $\text{Torr}$ ($O_2/Ar = 0.4$). The oxygen content in the films was changed by varying the oxygen pressure during the cooling procedure. The structural crystalline quality of the films was checked by x-ray diffraction analysis. $\theta − 2\theta$ and phi scans indicate an epitaxial growth with the $c$ axis oriented perpendicular to the substrate surface. Low angle x-ray reflectivity oscillations as well as finite size effects around the 001 peaks allow determination of the individual sample thickness, which was varied from 20 to 60 nm in this study. To prevent degradation during the lithographic processes, the films were protected by the subsequent in-situ deposition of an amorphous insulating layer. All the samples were photolithographically patterned using ion deposition of a 100 µm bridge wide. The relatively low thickness of the films allowed us to increase the measured voltages and to reduce the noise of the measurements. The $dc$ Hall voltage was measured using a three voltage contact technique and by adjusting the Hall voltage to zero in zero magnetic field. At each
temperature, the Hall voltage was measured by sweeping the magnetic field between $-6$ and $+6$ Tesla. During each measurement, the temperature, measured with a cerrom resistance, whose magnetoresistance is negligibly small, was stable within $\pm 5$ mK.

III. RESULTS AND DISCUSSION

Figure 3 shows the Hall constant $R_H$, as a function of temperature, for a series of GdBa$_2$Cu$_3$O$_{7-\delta}$ thin films. On Fig. 4, the curves correspond to films with critical temperatures of 84.6 K, 80.9 K, 79.0 K, 62.7 K, 48.9 K and 53.1 K. As can be seen, $T_c$ (Ref. 2) scales with the inverse Hall coefficient $R_H^{-1}$. Since $R_H^{-1}$ depends on the temperature, the relation between the doping level and $R_H^{-1}$ is not straightforward. Here, we will simply use, as other, the value of $R_H^{-1}$ at 100 K as a parameter related to the doping level. We find that the $T_c$ versus $R_H^{-1}(100$ K) curve obtained is in good agreement with YBa$_2$Cu$_3$O$_{7-\delta}$ single crystals data. This observation is consistent with a $T_c$ reduction and a change in doping in these thin films essentially related to a change in oxygen content. In Fig. 3 the highest $T_c$ sample has a resistivity which is typically twice the value found in optimally doped thin films. The value of $R_H^{-1}(100$ K) for this sample is slightly less than half the value found in optimally doped thin films, implying that in this thickness range, our films are not fully oxidized. One can also notice on Fig. 3 that the maximum in $R_H$ occurs in the normal state around 100 K for each sample and does not seem to be related to the $T_c$ of the sample. This point will be discussed below in detail.

In Fig. 2 the temperature dependence of $R_H^{-1}$ is shown for four representative samples. For the almost optimally doped sample with $T_c = 84.7$ K, $R_H^{-1}$ is linear in temperature over a wide temperature range as observed experimentally in optimally doped YBa$_2$Cu$_3$O$_{7-\delta}$ crystals. The temperature at which the inverse Hall constant deviates from linearity is shown by an arrow for each sample. For the highest $T_c$ sample the deviation in linearity is found to be very close to the peak in $R_H$ (minimum of $R_H^{-1}$). For more underdoped samples, the temperature at which the deviation occurs is shifted lower, the lower the $T_c$, the higher this temperature. These data are found to be consistent with the results of R. Jin and H. R. Ott, who made a systematic study of the Hall effect on YBa$_2$Cu$_3$O$_{7-\delta}$ single crystals. They found that the temperature at which $R_H^{-1}$ deviates from linearity is in good agreement with the temperature at which anomalies in the temperature dependence of other physical properties are reported, these anomalies being associated with the opening of the pseudogap at the temperature $T^*$.

We turn now to the analysis of the behavior of $\cot(\theta_H)$. Figure 3 shows for four selected samples ($\cot(\theta_H) - C)/T^2$ and $R_H$ as a function of temperature. Our data on the temperature dependence of $\cot(\theta_H)$ show, over a temperature range extending from 100 K to 300 K, a perfect $T^2$ dependence for all the doping levels investigated. No anomalies at temperatures close to $T^*$ (or at the temperature at which $R_H^{-1}(T)$ deviates from linearity) are found in $\cot(\theta_H)(T)$. As shown in Fig. 3 ($\cot(\theta_H) - C)/T^2$ is temperature independent for all the samples until a characteristic temperature at which a deviation is observed. We notice, as apparent on Fig. 3, that this temperature is essentially not affected by the sensitivity criterium used to define the deviation in the $T^2$ temperature dependence of $\cot(\theta_H)$. As can be seen also in Fig. 3, this second characteristic temperature $T'$, is well correlated with the temperature at which $R_H$ displays a peak and is almost independent of the doping level in the range investigated. At this temperature $T'$ no anomaly is observed in the resistivity, the peak in $R_H(\sim \rho_{xx}/\cot(\theta_H))$ has to be attributed to the change in $\cot(\theta_H)(T)$.

Since no deviation in $\cot(\theta_H)$ is observed at high temperature (close to $T'$), we conclude in the two relaxation times picture, that the deviation in $R_H^{-1} = \cot(\theta_H) \cdot H/\rho_{xx} \sim \tau_H^{-1} = \tau_r^{-1}$ has to be attributed to a change in $\tau_r^{-1}$, a diminution of the scattering rate related to the opening of the pseudogap. Since $\rho_{xx} \sim \tau_r^{-1}$, a deviation from the linear temperature dependence of the resistivity should be observed at the same characteristic temperature. A detailed analysis of the resistivity is difficult however, since, as often observed in underdoped thin films, the resistivity is not linear with temperature in this temperature range. It should be noticed that the $T^2$ temperature dependence of $\cot(\theta_H)$ is observed up to temperatures much higher than $T^*$ implying that the mechanism at the origin of the $T^2$ behavior extends at temperatures above the pseudogap phase.

In Fig. 4, we present a phase diagram $T$ versus $R_H^{-1}(100$ K) that summarizes the results of the paper. First $T_c$ versus $R_H^{-1}(100$ K) is plotted (squares) along with single crystals data (circles). The line is a guide to the eyes. As can be seen the critical temperatures of films and crystals fall on the same curve. The temperatures at which a deviation in the linear temperature dependence of $R_H^{-1}(T)$ is observed are indicated by triangles. These points follow the temperature dependence of the pseudogap temperature $T^*$ as discussed above. Finally, the diamonds are the well defined temperatures $T'$ at which a deviation in the $T^2$ behavior of $\cot(\theta_H)$ occurs (or at which $R_H$ is maximum). $T'$ falls between $T_c$ and $T^*$, clearly separating the pseudogap phase in two regions.

Before discussing this phase diagram and in particular the temperature $T'$, one can compare these results with very recent experiments on Bi$_2$Sr$_2$Ca$_{n-1}$Cu$_n$O$_y$ films which focusal on the doping dependence of the Hall response. In this system Z. Konstantinovic, Z. Z. Li and H. Raffy do not observe a correlation between the peak in $R_H$ and the deviation in $\cot(\theta_H)$. Their data, however, show that the temperature at which a deviation in $\cot(\theta_H)(T)$ occurs is much lower than $T^*$, in agreement
with the data presented here. We can also relate our data to other recent experimental findings. Analysis of the temperature dependence of \( R_H(T) \) have suggested that the peak in \( R_H(T) \) is related to the pseudogap. It is however clear from the experimental data shown in Fig. 4 that the temperature of the maximum in \( R_H(T) \) is well defined and does not follow the pseudogap temperature dependence, suggesting that the latter is not, at least directly, responsible for the behavior observed.

There have also been reports on \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta} \) and \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) compounds of the presence of vortex like excitations in the pseudogap phase. High frequency conductivity experiments and measurements of the Nernst effect have shown that vortex like excitations are found at temperatures up to 100 – 150 K. Although measured on different systems, these temperatures are above \( T_c \) but below the pseudogap temperature \( T^* \), as the characteristic temperatures \( T' \) measured in this work. Since experimentally without implying a particular scenario, the Hall angle has been shown to be sensitive to magnetic impurities through \( \tau_H^{-1} \), an interesting possibility is that vortex like excitations may affect the Hall angle. These vortex like excitations could be seen as a parallel, temperature dependent channel for magnetic scattering modifying the Hall angle and indirectly \( R_H \). The enhancement of \( \cot(\theta_H) \) due to this additional scattering source would be responsible, in this system for the break in the temperature dependence of \( R_H \).

The Nernst effect experiments, the high frequency experiments and the Hall data shown here point to a new crossover in the phase diagram. From an experimental point of view, three crossover temperatures are thus observed in underdoped cuprates: from weak \( (T_0) \) to strong pseudogap at \( T^* \) and, at lower temperature, the "new" crossover line discussed above. The crossover from weak to strong pseudogap has been intensively discussed in the literature theoretically and experimentally. Several theoretical approaches have been suggesting or calculating the existence of an additional structure in the strong pseudogap phase. In particular, phase diagrams with a new temperature \( T_{charge} \) at which charge ordering occurs have been discussed in the presence of charge inhomogeneities or stripes. Experimentally, T. Noda, H. Eisaki and S. Uchida have measured the Hall effect in \( \text{La}_{2-x-y}\text{Nd}_y\text{Sr}_x\text{CuO}_4 \) [Ref. 30] and have related the sharp decrease in \( R_H \) to one dimensional charge transport. We note also that recently, P. Devillard and J. Ranninger have predicted that there should exist a new characteristic temperature \( T^*_B \) in the pseudogap phase, between \( T^* \) and \( T_c \), that corresponds to the temperature at which the electron pairs, formed at higher temperature \( (T^*) \), become itinerant, defining an upper limit for partial Meissner screening (and vortex like excitations).

There are perhaps two main tentative theoretical frameworks for the pseudogap regime. In one, the pseudogap state is fluctuating superconductivity: the system is superconducting over short distances and time scales, but phase fluctuations destroy the long range superconducting order. The second main theoretical framework is related to the proximity of these systems to Mott insulators, and attempts to describe these strongly correlated systems with various schemes of spin charge separation or gauge theories. The Hall effect experiments presented in this paper raise in fact two related questions. Firstly whether the difference between the Hall and the resistivity temperature dependencies entails the existence of two different relaxation mechanisms and times, or whether it should be explained in a different way. If two different scattering times do indeed characterize the Hall effect situation in high temperature superconductors, then one must somehow reconcile this fact with the existence of the lower crossover. Unfortunately, the various spin charge separation theories, that provide in principle a natural explanation for the different relaxation times, have up to now, to our knowledge, not predicted a second crossover in the strong pseudogap regime. This difficulty could possibly be resolved if one assumes that the holons or chargons behave as 2D bosons: the new temperature \( T' \) could then be the critical temperature of the corresponding 3D system as recently discussed by Y. J. Uemura in the context of "preformed pairs". On the other hand, if the lower crossover is connected with the loss of short distance phase coherence between "preformed pairs", and therefore the disappearance of vortices, one is left in this phase fluctuation picture with the problem of finding a mechanism for the existence of two different relaxation times, and this, way above \( T^* \).

**IV. CONCLUSION**

In conclusion, we have studied the transport properties, resistivity and Hall effect, in a series of underdoped \( \text{GdBa}_2\text{Cu}_3\text{O}_{7-\delta} \) epitaxial thin films, grown by off-axis magnetron sputtering. We find that the inverse Hall constant is linear over a temperature range that decreases as \( T_c \) decreases. The temperature at which a deviation in \( R_H^{-1} \) occurs is close to the pseudogap temperature \( T^* \) measured in this material. We also find that \( \cot(\theta_H) \) displays a perfect \( T^2 \) temperature dependence over a large temperature range extending from about 100 K to 300 K. We observe for all the samples that the temperature at which the temperature dependence of the Hall angle deviates from the \( T^2 \) behavior is precisely correlated to the temperature at which \( R_H \) displays a peak. This characteristic temperature, found to lie between \( T_c \) and the pseudogap temperature \( T^* \), does not depend markedly on the doping level and defines a new crossover in the phase diagram.

We thank Z. Tešanović for bringing to our attention the possible importance of vortices in the pseudogap phase, D. Marré for help in the initial stage of this project and D. Chablaix for technical help. This work was supported
The pseudogap is directly observed in STM experiments, Ch. Renner, B. Revaz, J.-Y. Genoud, K. Kadowaki, and Ø Fischer, Phys. Rev. Lett. 80, 149 (1998), and in ARPES measurements, H. Ding, T. Yokoya, J. C. Campuzano, T. Takahashi, M. Randeria, T. Mochiku, K. Kadowaki, and J. Giapintzakis, Nature 382, 51 (1996), and A. G. Loeser, Z.-X Shen, D. S. Dessau, D. S. Marshall, C. H. Park, P. Fournier, and A. Kapitulnik, Science 273, 325 (1996).

V. J. Emery and S. A. Kivelson, Nature 374, 434 (1995).

P. Devillard and J. Ranninger, Phys. Rev. Lett. 84, 5200 (2000).

V. J. Emery, E. Fradkin, S. A. Kivelson, and T. C. Lubensky, Phys. Rev. Lett. 85, 2160 (2000).

M. Vojta, Y. Zhang, and S. Sachdev, cond-mat/0007170.

S. Chakravarty, R. B. Laughlin, D. K. Morr, and C. Nayak, cond-mat/0005413.

Y. J. Uemura, cond-mat/0012016.

Y. Abe, K. Segawa, and Y. Ando, Phys. Rev. B 60, R15055 (1999).

Z. Konstantinović, Z. Z. Li, and H. Raffy, Phys. Rev. B 62, R11989 (2000).

T. Noda, H. Eisaki, and S. Uchida, Science 286, 265 (1999).

H. A. Mook, P. Dai, F. Doğan, and R. D. Hunt, Nature 404, 729 (2000).

J. Corson, R. Mallozzi, J. Orenstein, J. N. Eckstein, and I. Bozovic, Nature 398, 221 (1999).

Z. A. Xu, N. P. Ong, Y. Wang, T. Kakeshita, and S. Uchida, Nature 406, 486 (2000).

J.-M. Triscone and Ø. Fischer, Rep. Prog. Phys. 60, 1673 (1997).

Although for similar cooling procedures, the $T_c$ of samples with different thicknesses were different, we found that the $T_c$ versus $R_H^2(100K)$ data points fall on the same curve, irrespective of the film thickness.

$T_c$ is defined as the temperature at which the resistivity, on a logarithmic scale, falls in the noise level of the measurements.

R. Jin and H. R. Ott, Phys. Rev. B 57, 13872 (1998).

T. Ito, K. Takenaka, and S. Uchida, Phys. Rev. Lett. 70, 3995 (1993).

A. Carrington, D. J. C. Walker, A. P. Mackenzie, and J. R. Cooper, Phys. Rev. B 48, 13051 (1993).

The difficulty to fully oxidize these samples might be related to strains in very thin films.

N. P. Ong, in Physical Properties of High Temperature Superconductors (edited by D. M. Ginsberg, World Scientific, Singapore, 1990), vol. 2, p. 459.

The influence of the pseudogap on the Hall constant in YBa$_2$Cu$_4$O$_8$ and in La$_{2-x}$Sr$_x$CuO$_4$ is respectively discussed in Ref. 39 and Ref. 40.

In the experiment of T. R. Chien et al., adding zinc (modifying the in plane magnetic scattering) shifts cot($\theta_H$) at higher values, the change in $C$ being directly proportional to the amount of Zn.

This particular compound has a structural phase transition at a temperature $T_0$ and the low temperature phase stabilizes charges inhomogeneities (stripes).

P. A. Lee, Physica C 317, 194 (1999).

T. Senthil and M. P. A. Fisher, cond-mat/9912380.

B. Bucher, P. Steiner, J. Karpinski, E. Kaldis, and P. Wachter, Phys. Rev. Lett. 70, 2088 (1993).

T. Senthil and M. P. A. Fisher, cond-mat/9912380.

H. Y. Hwang, B. Batlogg, H. Takagi, H. L. Kao, J. Kwo, R. J. Cava, J. J. Krajewski, and W. F. Peck, Jr., Phys. Rev. Lett. 72, 2636 (1994).
FIG. 1. Temperature dependence of the Hall constant $R_H$ for a series of GdBa$_2$Cu$_3$O$_{7−δ}$ thin films. The curves correspond to films with critical temperatures of 84.6 K (♦), 80.9 K (X), 79.0 K (+), 62.7 K (▲), 48.9 K (■) and 53.1 K (▼).

FIG. 2. Temperature dependence of $R_H^{-1}$ for four representative samples. The temperature at which the inverse Hall constant deviates from the linearity is shown by an arrow for each sample. This characteristic temperature can be associated with the pseudogap temperature $T^*$. 
FIG. 3. \((\cot(\theta_H) - C)/T^2\) (right axis) and \(R_H\) (left axis) for four selected samples are shown as a function of temperature. \((\cot(\theta_H) - C)/T^2\) is temperature independent for all the samples until a characteristic temperature \(T'\) which is well correlated with the temperature at which \(R_H\) displays a peak.

FIG. 4. The phase diagram \(T\) versus \(R_H^{-1}(100\,\text{K})\) (logscale) summarizes the results of the paper. \(T_c\) versus \(R_H^{-1}(100\,\text{K})\) (■), along with single crystals data from Ito et al. (●), the temperature at which \(R_H^{-1}(T)\) deviates from linearity associated to the pseudogap temperature \(T^*\) (▲) and \(T'\) (♦), the temperature at which a deviation in the \(T^2\) behavior of \(\cot(\theta_H)\) is observed, are plotted.