

## Hall effect of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ single crystals

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For a number of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals with various oxygen concentrations and differing values of the zero magnetic-field critical temperature  $T_{c0}$ , the Hall effect has been investigated between 10 and 300 K by applying magnetic fields  $\mathbf{H} \parallel c$  and currents  $\mathbf{I} \perp c$ , where  $c$  denotes the direction perpendicular to the Cu-O planes of this material. These measurements yield that the sign change of the Hall voltage close to  $T_{c0}$  varies with  $\delta$  and eventually disappears in heavily oxygen-depleted  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals. Such behavior can qualitatively be explained by a model calculation based on the time-dependent Ginsburg-Landau theory. In the normal state, we find that the Hall coefficient  $R_H$  and the Hall angle  $\theta_H$  vary as  $R_H^{-1} \propto T$  and  $\cot \theta_H \propto T^2$  in an extended temperature range, as has previously been found for fully oxygenated  $\text{YBa}_2\text{Cu}_3\text{O}_7$  and  $\text{YBa}_2\text{Cu}_{3-y}\text{M}_y\text{O}_7$  ( $M = \text{Ni}, \text{Zn}$ ). Deviations from these behaviors for both  $R_H^{-1}$  and  $\cot \theta_H$  are observed below a characteristic temperature  $T^*$ , whereby  $T^*$  increases with decreasing  $T_{c0}$ , i.e., increasing oxygen depletion. This implies a temperature-induced variation of the normal-state electronic spectrum and/or the scattering parameters governing the electronic transport of this material.

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### I. INTRODUCTION

The Hall effect of high- $T_c$  cuprates has been studied extensively in both the mixed and the normal state. Numerous experiments with nearly optimally doped cuprates<sup>1,2</sup> have shown quite generally that the inverse of the Hall coefficient  $R_H^{-1}$  varies linearly with temperature in the normal state but changes its sign at temperatures in the vicinity of the zero-field superconducting transition temperature  $T_{c0}$ . Recent studies on  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (Ref. 3) and  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (Refs. 4–6) showed that at doping levels away from optimal conditions, more or less pronounced deviations of  $R_H(T)$  from the above sketched behavior are observed at temperatures above and below  $T_{c0}$ . This is of importance because the above-mentioned temperature variations of transport properties have been matched with theoretical predictions aiming at characterizing the unusual nature of these conducting cuprates. Additional measurements have demonstrated that for hole-type cuprates the magnitude of the negative Hall conductivity close to  $T_{c0}$  increases with increasing pinning strength,<sup>7–9</sup> suggesting that the sign change of the Hall voltage is also related to pinning-induced backflow of charge carriers.<sup>10</sup> Furthermore, the normal-state Hall effect<sup>11,12</sup> and NMR measurements<sup>13</sup> on Zn-doped  $\text{YBa}_2\text{Cu}_{3-y}\text{Zn}_y\text{O}_{7-\delta}$  suggest that Zn doping only creates in-plane disorder without affecting the temperature dependences of the transport coefficients or the carrier density, although the value of  $T_{c0}$  distinctly decreases with increasing Zn content. In a more recent report,<sup>14</sup> it was claimed that varying the oxygen content of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  material is roughly equivalent to substituting Zn for Cu and it was argued that Zn or oxygen doping may not affect the normal-state spin-fluctuation spectrum of  $\text{YBa}_2\text{Cu}_{3-y}\text{Zn}_y\text{O}_{7-\delta}$ . All these controversial experimental results and their interpretation indicate that both the experimental and the theoretical situations regarding the influence of doping on the Hall effect are not settled and the origin of

the anomalous Hall response in both the mixed and the normal state of a given superconductor is not fully understood.

In an attempt to enhance the data base for improving the understanding of the Hall anomalies and their relation with the doping level, we have measured the Hall effect on a number of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals with differing oxygen concentrations and hence differing values of the critical temperature  $T_{c0}$  between 39 and 91 K. Our experimental results reveal a general trend in the temperature variations of the Hall parameters  $R_H$  and  $\theta_H$  in both the mixed and normal state upon changing the oxygen content. We compare our data and results of our analysis with theoretical models<sup>15–19</sup> and other similar or related experimental findings,<sup>3,12,20–22</sup> and we identify both agreements and disagreements with respect to observations of anomalous features of the Hall effect in the mixed and the normal state of underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  in this and other work.

### II. SAMPLES AND EXPERIMENT

Using the technique described in Ref. 2, the in-plane longitudinal voltage  $V_{xx}$  and the in-plane Hall voltage  $V_{xy}$  of seven monocrystalline  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  samples have been measured. The dc current  $\mathbf{I}$  and the magnetic field  $\mathbf{H}$  were applied perpendicular and parallel to the  $c$  axis of the crystals, respectively. In this study, we selected a  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystal with  $T_{c0} = 90.5$  K, which was grown by the method described in detail elsewhere.<sup>23</sup> In order to adjust the oxygen content, the single crystal was cut into three pieces. One of them was labeled as sample I with the dimensions of  $2.2 \times 1.0 \times 0.2$  mm<sup>3</sup>. Of the two others, one ( $1.0 \times 0.5 \times 0.2$  mm<sup>3</sup>) was annealed in a tube furnace in flowing  $\text{O}_2$ , and the third ( $0.5 \times 0.4 \times 0.1$  mm<sup>3</sup>) was annealed in air. This resulted in  $T_{c0} = 91.0$  K for sample II and 84.0 K for sample III, respectively. By subsequent annealing of sample I at 350 °C in Ar atmosphere and altering the annealing time, we varied the  $T_{c0}$  values of the same piece of crystal. This pro-

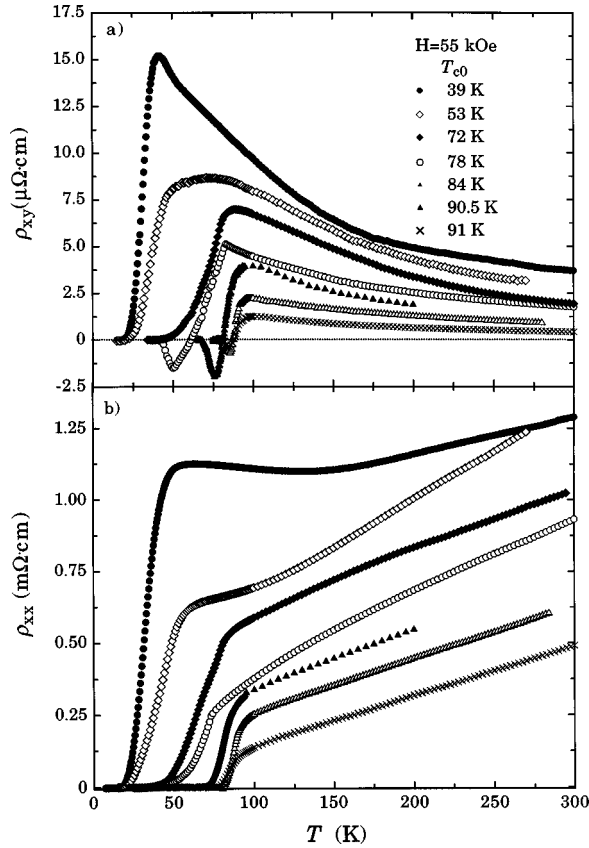


FIG. 1. Temperature dependences of the in-plane Hall resistivity  $\rho_{xy}$  (a) and longitudinal resistivity  $\rho_{xx}$  (b) between 10 and 300 K for samples I–VII. All data were taken at  $H=55$  kOe.

cess lead to sample IV with a  $T_{c0}$  of 78.0 K and samples V, VI, and VII with  $T_{c0}=72.0$ , 53.0, and 39.0 K, respectively. The annealing process indicates that the increase/decrease of  $T_{c0}$  is due to the increase/decrease of the oxygen content of the sample, i.e., an enhancement of the oxygen deficiency  $\delta$  results, as is long well-known, in a reduction of  $T_{c0}$ . For ensuring consistency,  $T_{c0}$  for all samples studied was defined as the highest temperature at which the zero-field longitudinal resistivity  $\rho_{xx}(H=0)$  is zero within experimental resolution.

### III. RESULTS AND DISCUSSION

#### A. Superconducting state and sign reversal of the Hall voltage

In Fig. 1(a), we display the temperature dependence of the Hall resistivity  $\rho_{xy}$  between 10 and 300 K for seven  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  samples. The longitudinal resistivity  $\rho_{xx}$  in the same temperature interval is shown in Fig. 1(b). For easier comparison, all these data were taken at  $H=55$  kOe. It may be seen in Fig. 1(a) that, without exception, at high temperatures  $\rho_{xy}$  is positive and increases with decreasing  $T$ . All the  $\rho_{xy}$  vs  $T$  curves are shifted upwards as  $\delta$  increases, i.e., the doping and  $T_{c0}$  decreases. This is consistent with previous results observed for oxygen-depleted  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals<sup>24</sup> and films,<sup>5</sup> which established that the decrease of  $T_{c0}$  is caused by a decrease of the hole concentration. At temperatures approaching the respective  $T_{c0}$  from above,  $\rho_{xy}$  for all samples gradually decreases with decreasing  $T$ . Even-

tually,  $\rho_{xy}$  for samples I–IV changes sign. Below the negative maximum,  $\rho_{xy}$  again increases and finally essentially vanishes. For samples V–VII,  $\rho_{xy}$  continuously decreases to zero without experiencing the sign reversal around  $T_{c0}$ . Our data shown in Fig. 1(a) thus indicate that the negative Hall resistivity at  $H=55$  kOe is only observed for samples I–IV with values of  $T_{c0}$  exceeding 72 K. We further note that the amplitude of the negative maximum  $|\rho_{xy}^{\min}|$  also depends on the oxygen content. Starting from  $T_{c0}$  of the optimally doped material,  $|\rho_{xy}^{\min}|$  initially increases with decreasing  $T_{c0}$  then decreases and eventually disappears. This confirms the data reported in Ref. 6, where  $\rho_{xy}$  displays no negative anomaly for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals with  $T_{c0} \leq 75$  K at any applied field. A very similar behavior is also observed in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ , if the doping is varied by changing the value of  $x$ ,<sup>3</sup> suggesting that quite generally the Hall response of high- $T_c$  cuprates in both the superconducting and normal state strongly depends on the doping level.

Concerning the issue of the sign reversal of the Hall resistivity in cuprate superconductors, the models for its explanation that have been proposed so far have not given an exhaustive description, although numerous attempts have been made to explain this unexpected phenomenon. It has been suggested, for instance, that the flux pinning in the mixed state may induce a backflow of charge carriers and therefore cause a negative contribution to the Hall voltage.<sup>10</sup> For  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , it seems established that the pinning potential increases with increasing  $\delta$ , i.e., decreasing  $T_{c0}$ .<sup>25</sup> Therefore, if the negative Hall resistivity simply arises from flux pinning, one expects that  $|\rho_{xy}^{\min}|$  increases with decreasing  $T_{c0}$ . Our data reveal a nonmonotonic dependence of  $|\rho_{xy}^{\min}|$  on  $\delta$ , indicating that the pinning strength cannot be the only reason for the negative Hall anomaly and confirming previous arguments in Ref. 15.

Another explanation of the origin of the sign change of the Hall resistivity involves a two-band scenario with both electron- and hole-type carriers.<sup>26,27</sup> According to Ref. 26, the electron and hole carriers respond differently to variations of the temperature and, naturally, to the magnetic field, and hence the sign of the Hall resistivity is determined by the competition of these two contributions. Based on this picture, the sign reversal of the Hall resistivity is expected to be more pronounced if the hole-carrier concentration of a hole-type superconducting cuprate like  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is reduced. Since the decrease of oxygen content in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is generally considered to lower the hole concentration, our experimental result is obviously not consistent with this two-band model over the whole concentration range.

According to our experimental observation, there is no doubt that the sign change of the Hall resistivity is, at least partially, controlled by the doping level. A model calculation, based on the time-dependent Ginsburg-Landau equation, reveals that the Hall effect in the superconducting fluctuation regime may be explained as a result of the electron-hole asymmetry near the Fermi level and that the sign of Hall conductivity is determined by the sign of  $\partial N(0)/\partial \mu$  (Ref. 28) or  $\partial \ln T_c / \partial \ln \mu$ ,<sup>29</sup> where  $N(0)$  is the density of electronic states at the Fermi level and  $\mu$  is the chemical potential. Several theoretical groups<sup>15–17</sup> found that the Hall conductivity in the fluctuation regime, both above and below

$T_{c0}$ , is the sum of two contributions. Depending on the temperature regime, the total Hall conductivity  $\sigma_{xy}$  may thus be written as

$$\sigma_{xy} = \sigma_{xy}^{\text{fl}} + \sigma_{xy}^n, \quad T > T_{c0}, \quad (1a)$$

$$\sigma_{xy} = \sigma_{xy}^s + \sigma_{xy}^{\text{qp}} = C_1/H + C_2 \cdot H, \quad T < T_{c0}. \quad (1b)$$

The first terms on the right-hand side of Eqs. (1a) and (1b) are due to the electron-hole asymmetry near the Fermi level. The second terms represent the normal-state Hall conductivity which, below  $T_{c0}$ , applies for the quasiparticles in the vortex cores. The parameters  $C_1$  and  $C_2$  are field-independent constants.<sup>15,16</sup> In Refs. 15, 17, and 30, the motion of quasiparticles inside the vortex cores has been treated by regarding a cylinder of material in its normal state, implying that  $\sigma_{xy}^{\text{qp}}$  is essentially given by  $\sigma_{xy}^n$ . The signs of the fluctuation-contributed Hall conductivity  $\sigma_{xy}^{\text{fl}}$  above  $T_{c0}$  and the Hall conductivity  $\sigma_{xy}^s$  in the mixed-state are controlled by the imaginary part  $\gamma''$  of the order-parameter relaxation time, which is related to  $\partial N(0)/\partial \mu$ .<sup>16</sup> The Hall-voltage measurements on nearly optimally doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (Refs. 24 and 31) and Bi-based superconducting cuprates<sup>32</sup> have shown that the experimental data at temperatures close above and below  $T_{c0}$  can be well described by Eq. (1b), except in the low-field regime. The decomposition of these experimental data using Eq. (1b) confirms that  $C_1$  is negative and, as predicted theoretically for hole-doped cuprate superconductors,  $C_2$  is positive. The negative sign of  $C_1$  eventually leads to the sign change of the total Hall conductivity. In view of the above-mentioned model calculations,<sup>15-17</sup> the negative sign of  $C_1$  implies that  $\gamma'' > 0$  or  $\partial N(0)/\partial \mu > 0$ . Recently, a systematic study of the Hall conductivity in  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  (Ref. 3) has demonstrated that, below  $T_{c0}$ ,  $C_2$  is always positive and increases monotonically with increasing  $x$  at a fixed  $T/T_{c0} < 1$ . However, the sign of  $C_1$  depends on the doping level  $x$ . It is negative in the under- and slightly overdoped region but is positive in the heavily overdoped region. The magnitude of  $C_1$  varies nonmonotonically with  $x$ . With increasing  $x$ ,  $C_1$  initially decreases then increases again.<sup>3</sup>

In Fig. 2, we emphasize the temperature dependence of the Hall conductivity  $\sigma_{xy} \approx \rho_{xy}/\rho_{xx}^2$  of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  in a fixed magnetic field close to  $T_{c0}$ . In order to facilitate the comparison,  $\sigma_{xy}$  is plotted as a function of  $T/T_{c0}$  between 0.8 and 1.2. It may be noted that, above about  $1.05T_{c0}$ ,  $\sigma_{xy}$  is positive for all samples and slightly increases with decreasing  $T$ . Below  $1.05T_{c0}$ ,  $\sigma_{xy}$  for samples V–VII stays positive and continues to increase with decreasing  $T$ . In contrast,  $\sigma_{xy}$  for samples I–IV decreases and changes sign as the temperature is reduced. In these cases, the negative contribution to the Hall conductivity is large enough to dominate the total Hall conductivity just below  $T_{c0}$ . Considering the above-mentioned two-component model<sup>15-17</sup> and previous experimental results,<sup>2,3,6,24,31,32</sup> the dominant negative contribution to  $\sigma_{xy}$  below  $T_{c0}$  ought to be attributed to the mixed-state Hall conductivity  $\sigma_{xy}^s$ . At a fixed value of  $T/T_{c0} < 1$ , Fig. 2 shows that  $\sigma_{xy}$  generally decreases from positive to negative then increases with increasing  $T_{c0}$  except for sample VII. Since  $\sigma_{xy}^n$  increases with increasing hole (oxygen) concentration in the normal state (see Fig. 2) and  $\sigma_{xy}$  varies smoothly through the superconducting transition, we

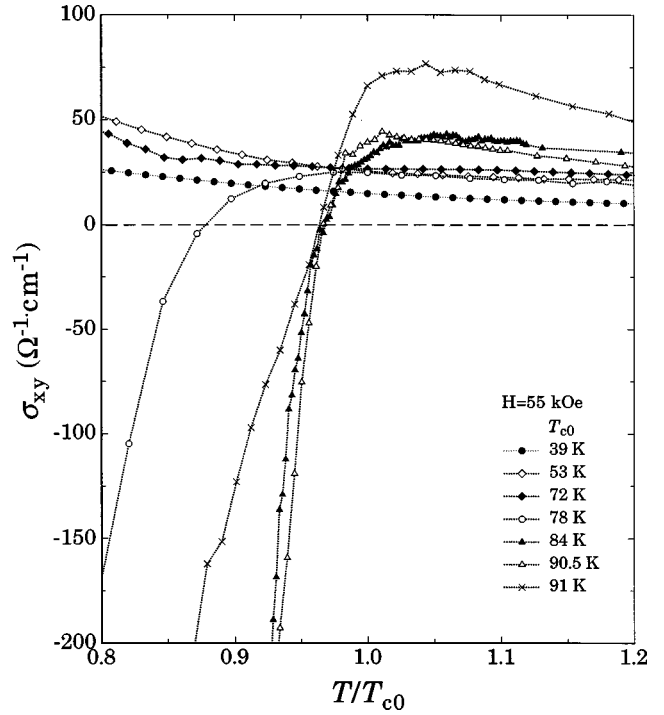


FIG. 2. Temperature dependence of the Hall conductivity  $\sigma_{xy}$  as a function of  $T/T_{c0}$  between 0.8 and 1.2 for samples I–VII. All data were taken at  $H = 55$  kOe. The dotted lines are to guide the eye.

may expect the same general variation of  $\sigma_{xy}^{\text{qp}}$  in the mixed state. This was indeed verified for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .<sup>3</sup> Our data shown in Fig. 2 thus imply that, at a fixed relative temperature  $T/T_{c0} < 1$ , the magnitude of the negative  $\sigma_{xy}^s$  increases with increasing  $T_{c0}$  except for sample II with the highest value of  $T_{c0}$ . Thus the maximum negative contribution to  $\sigma_{xy}$  is found for nearly optimally doped material, consistent with the results obtained for  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .<sup>3</sup> This confirms that also for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$   $\sigma_{xy}^s$  depends sensitively on the oxygen concentration.

## B. Normal-state Hall effect

One way to discuss the normal-state Hall response of high- $T_c$  cuprates is offered by comparing the experimental data with the theoretical model proposed by Anderson.<sup>33</sup> In this model, the temperature dependence of the Hall coefficient  $R_H$  in the normal state is predicted to vary as

$$R_H^{-1} = AT + B, \quad T > T_{c0}. \quad (2)$$

Here,  $A$  and  $B$  are temperature-independent constants. It has been found that Eq. (2) describes the experimental data quite well over a wide temperature range for the optimally doped high- $T_c$  materials. In order to test the validity of Eq. (2) for underdoped materials, we have converted our data shown in Fig. 1(a) into  $R_H^{-1} = H/\rho_{xy}$ . In Fig. 3, we have plotted the temperature dependences of  $R_H^{-1}$  for samples I–VII. At all temperatures between  $T_{c0}$  and 300 K, the magnitude of  $R_H^{-1}$  decreases with increasing oxygen depletion. For all samples,  $R_H^{-1}$  varies approximately linearly with  $T$  at temperatures exceeding a characteristic temperature  $T^*(\delta)$ , which increases with decreasing oxygen concentration. Hence for

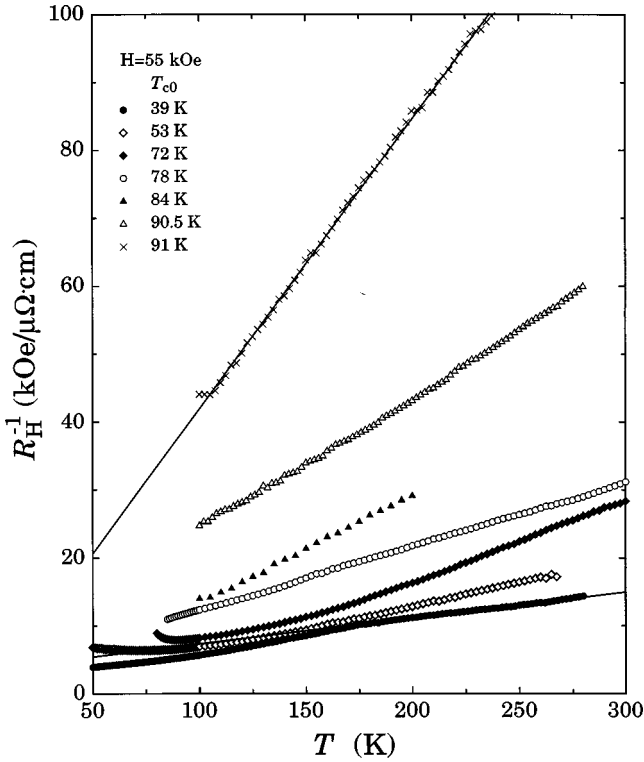


FIG. 3. Temperature dependence of the inverse Hall coefficient  $R_H^{-1}$  between  $T_{c0}$  and 300 K for samples I–VII. The solid lines are fits to experimental data above  $T^*$  (see text) using Eq. (2) for sample II and VII, respectively.

$T > T^*$ , the  $R_H^{-1}$  vs  $T$  data may be fitted with Eq. (2) and we find that the corresponding slope  $A$  decreases from 0.43 to 0.04 kOe/ $\mu\Omega$  cm K as  $T_{c0}$  decreases from 91 to 39 K except for sample IV. In contrast, the parameter  $B$  generally increases with decreasing  $T_{c0}$ . Thus the suppression of  $T_{c0}$  results in a suppression of the ‘Hall coefficient slope’  $A$  and an enhancement of  $B$ . Similar correlations were observed in Zn-doped  $\text{YBa}_2\text{Cu}_3-y\text{Zn}_y\text{O}_7$  (Ref. 11) and oxygen-deficient  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  thin films.<sup>34</sup>

For our samples,  $R_H^{-1}$  gradually deviates from the linear  $T$  dependence below  $T^*$ . As may be seen in the inset of Fig. 5, to be discussed later, the value of  $T^*$  systematically increases with decreasing  $T_{c0}$ . This is in conflict with the results presented in Refs. 12 and 14, where  $T^*$  does not change with Zn or O doping in  $\text{YBa}_2\text{Cu}_3-y\text{Zn}_y\text{O}_{7-\delta}$ , although  $T_{c0}$  rapidly decreases with increasing  $y$  or  $\delta$ .

Anderson’s model<sup>33</sup> is also predictive with respect to the temperature dependence of the Hall angle. The relevant equation is

$$\cot \theta_H = \alpha T^2 + \beta, \quad (3)$$

where  $\alpha$  and the impurity-induced contribution  $\beta$  are temperature-independent constants. The Hall angle  $\theta_H$  may be obtained from  $\cot \theta_H = \rho_{xx}/\rho_{xy}$ . In Fig. 4, we show plots of  $\cot \theta_H$  versus  $T^2$  for samples I–VII. For each sample, the data fall on an approximately straight line in the high-temperature range and hence our experimental data may be fitted by using Eq. (3) at high temperatures. As expected, we find that both  $\alpha$  and  $\beta$  are sample dependent. We confirm the previously demonstrated trend<sup>11,12</sup> that the value of  $\beta$  in-

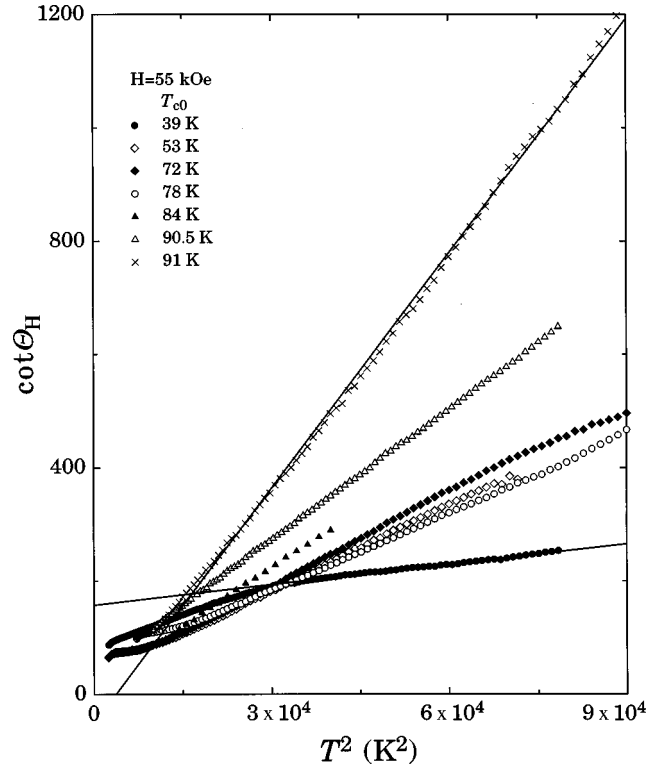


FIG. 4. Temperature dependence of the Hall angle  $\cot \theta_H$  between  $T_{c0}$  and 300 K for samples I–VII. The solid lines are fits to experimental data above  $T^*$  using Eq. (3) for sample II and VII, respectively.

creases with decreasing  $T_{c0}$ , i.e., with increasing oxygen deficiency  $\delta$ , except for sample IV. The value of  $\alpha$ , however, shows an opposite tendency, consistent with the observation on thin films of oxygen-deficient  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (Refs. 5, 34) and underdoped  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ .<sup>35</sup> We recall that, for Zn-doped  $\text{YBa}_2\text{Cu}_3-y\text{Zn}_y\text{O}_{7-\delta}$ , a different  $\alpha$  variation with  $T_{c0}$  was observed.<sup>11</sup>

According to the mentioned model calculation,<sup>11,33</sup> the coefficient  $\alpha$  is proportional to the carrier density  $n$ . Our data thus confirm that the decrease of oxygen content (decreasing  $T_{c0}$ ) not only enhances the defect-scattering manifested by an increasing value of  $\beta$ , but also reduces the carrier density  $n$  (decreasing  $\alpha$ ), which coincides with the decrease of  $R_H^{-1}(T)$  upon oxygen depletion in the normal state.

Our data and previous observations<sup>5,11,34</sup> provide support for the validity of Eq. (3) in a restricted temperature range which by itself is material dependent. Similar as in the case of  $R_H^{-1}(T)$ , we note a characteristic temperature  $T^*$ , below which  $\cot \theta_H$  deviates from the simple  $T^2$  dependence. Again,  $T^*$  increases as  $T_{c0}$  decreases and we note that the  $T^*$  values from both the  $R_H^{-1}(T)$  and  $\cot \theta_H(T)$  plots coincide within some uncertainty limits (see the inset of Fig. 5). This suggests that the value of  $T^*$  is simply a function of the doping level. Thus, in Fig. 5, we have plotted the sample dependence of  $T^*$  obtained from both  $R_H^{-1}(T)$  (crosses) and  $\cot \theta_H(T)$  (open circles) curves as a function of  $R_H^{-1}(200 \text{ K})$ , which may be taken as representing the carrier concentration of the investigated specimens. For comparison, the corresponding values of  $T_{c0}$  vs  $R_H^{-1}(200 \text{ K})$  (solid circles) are included in Fig. 5. The plots in Fig. 5 clearly

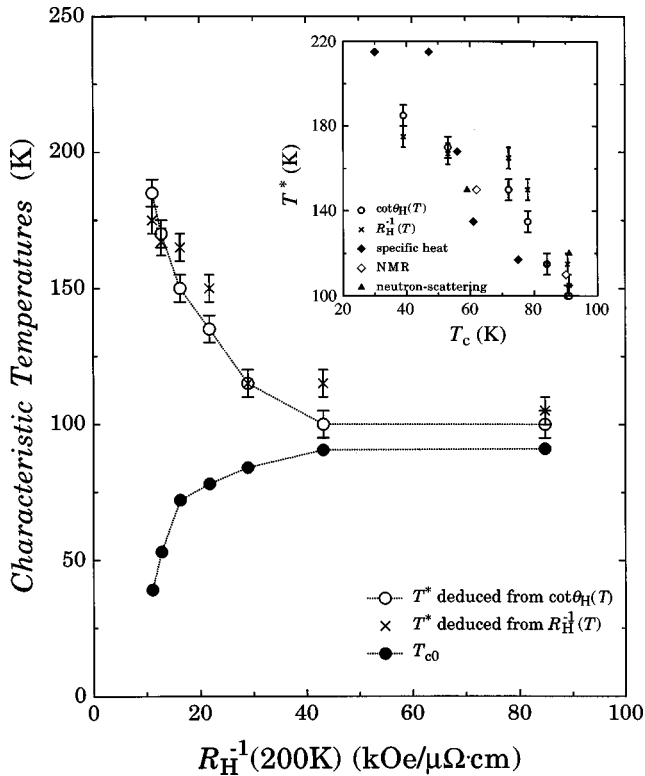


FIG. 5. Characteristic temperature  $T^*$  deduced from both  $R_H^{-1}(T)$  (crosses) and  $\cot \theta_H(T)$  (open circles) (see text) and the zero-field superconducting transition temperature  $T_{c0}$  as a function of  $R_H^{-1}(200\text{ K})$  (solid circles). The dotted lines are to guide the eye. The inset shows the dependence of  $T^*$  on  $T_{c0}$  obtained from  $R_H^{-1}(T)$  (crosses),  $\cot \theta_H(T)$  (open circles),  $\rho_{xx}(T)$  (open triangles), electronic specific heat (solid diamonds), NMR (empty diamonds), and neutron scattering (solid triangles).

demonstrate that for low doping the distinct decrease of  $T_{c0}$  with increasing  $\delta$  is complemented by an equally strong enhancement of  $T^*$ . With increasing  $R_H^{-1}(T)$  or doping,  $T_{c0}(\delta)$  and  $T^*(\delta)$  gradually approach each other and finally coincide for nearly optimally doped material. This result is in very good agreement with a previous report in Ref. 4.

In order to relate  $T^*(\delta)$  deduced from these Hall-effect measurements with other experimental observations concerning underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , we consider anomalies in the temperature dependences of other physical properties that have been reported in the literature and presented in this paper. In view of previous theoretical<sup>19,36,37</sup> and experimental work,<sup>4,21,22,38</sup> the most plausible and most frequently discussed scenario for causing the anomalous normal-state physical properties of cuprates is the scattering of charge carriers by spin fluctuations in the Cu-O plane. Both NMR (Ref. 21) and neutron-scattering<sup>22</sup> (NS) measurements suggest that antiferromagnetic (AF) correlations lead to a spin gap in the normal state of underdoped superconducting cuprates. For  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , Loram and co-workers<sup>20</sup> claim to have found that the electronic specific-heat parameter  $\gamma$  is temperature dependent and exhibits a broad maximum at temperatures above  $T_{c0}$  for underdoped material. This maximum is seen to shift to lower temperatures with increasing oxygen content. The results obtained from NMR and inelastic neutron-scattering experiments have also been interpreted

as to reveal the onset of a gap formation in the spin-excitation spectrum of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  with varying oxygen concentration.<sup>21,22</sup> Likewise, measurements of the in-plane electrical resistivity,<sup>4,5</sup> optical spectroscopy,<sup>39</sup> and Raman-scattering<sup>40</sup> experiments reveal anomalies in the normal state of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  samples. As may be seen in Fig. 1(b), the temperature variation of our  $\rho_{xx}$  data is close to linear for all samples at high temperatures. Deviations from this behavior are again observed at temperatures below  $T^*$ . For samples I–V,  $\rho_{xx}(T)$  decreases faster below  $T^*$  than above. This is not true for the two samples with the lowest values of  $T_{c0}$ . The trend towards an increasing  $\rho_{xx}$  with decreasing  $T$  is most likely due to an interfering contribution of the  $c$ -axis resistivity, which is increasingly nonmetallic in the normal state of heavily underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ .<sup>4</sup> Our data confirm the previously published claim<sup>4,5</sup> of an increase of  $T^*$ , as deduced from  $\rho_{xx}(T)$ , with decreasing oxygen content, i.e.,  $T_{c0}$ . The comparison of our  $T^*$  values with those derived from the unusual normal-state behavior of the electronic specific heat, NMR data, and results of neutron-scattering experiments are shown in the inset of Fig. 5. We note that the values of  $T^*$  obtained from a variety of investigations on many different samples show the same general trend of increase with decreasing doping level. This suggests that all the reported anomalies of the normal state are of the same origin, depending on the oxygen doping level in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ .

Theoretically, Nagaosa and Lee<sup>36</sup> predict that the pair-formation temperature  $T_D$  of spinons and the spin pseudogap  $\Delta_s \propto \sqrt{|T - T_D|}$  decrease with increasing hole concentration, and that  $T_D > T_{c0}$  for underdoped materials. Thus, we expect that the spin gap, for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , decreases with increasing oxygen concentration and finally disappears for fully oxygenated material. As mentioned above, evaluations of the electronic specific heat,<sup>20</sup> and observations in NMR,<sup>21</sup> Raman,<sup>40</sup> and optical conductivity measurements<sup>39</sup> have demonstrated this trend. In view of the evidence accumulated in the present study, we conclude that also the Hall-effect data provide further support for this view. For nearly optimally doped samples,  $T^*$  virtually coincides with  $T_{c0}$  and in this case the deviations of  $R_H(T)$  and  $\cot \theta_H(T)$  are most likely due to superconducting fluctuation effects that are taken into account by Eq. (1a) and have been discussed earlier.<sup>2,15</sup> Whether  $T^*$  really marks the onset of a spinon-pair formation cannot unambiguously be concluded from the results presented above.

#### IV. SUMMARY AND CONCLUSION

In summary, we have made a systematic study of the Hall effect of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  single crystals with varying carrier concentrations and hence different values of  $T_{c0}$ . The quantities related with the Hall effect are very sensitive to the oxygen content in both the mixed and the normal state. The sign of the Hall conductivity below  $T_{c0}$  is most likely determined by details of the electronic structure at the Fermi level, which obviously varies with the itinerant carrier concentration. In the normal state, both  $R_H^{-1}(T)$  and  $\cot \theta_H(T)$  show simple temperature dependences at high temperatures but deviate considerably from this behavior below  $T^*$ , whereby  $T^*$  increases with decreasing oxygen content. The

$T^*$  values derived from these Hall data are consistent with crossover temperatures that have been evaluated from other normal-state properties of underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  and have been claimed to mark the onset of a gap for spin excitations.

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