Hall effect and magnetoresistance in $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ films

P. Seng, J. Diehl, S. Klimm, S. Horn, R. Tidecks, and K. Samwer
Institut für Physik, Universität Augsburg, Memminger Strasse 6, D-86135 Augsburg, Germany

H. Hänsel and R. Gross
Lehrstuhl für Experimentalphysik II, Universität Tübingen, Morgenstelle 14, D-72076 Tübingen, Germany

(Received 2 December 1994; revised manuscript received 3 April 1995)

The electrical transport properties of the cuprate superconductor $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ have been studied in the mixed state and in the normal conducting state by measuring the Hall voltage and the longitudinal voltage of an epitaxial thin film. A broadening of the resistive transition with increasing fixed magnetic field and a sign reversal of the Hall effect in the mixed state were observed. In the normal state, the zero-field resistivity shows a quadratic temperature dependence. Magnetoresistance measurements for $T=102$ and 55 K yield a B^2 dependence up to $B=10$ T without any saturation tendency. The normal-state Hall coefficient is temperature dependent with a sign change from negative to positive for decreasing temperature. The data can be interpreted within a two-band model.

I. INTRODUCTION

In 1989 Tokura *et al.*[1] and Takagi *et al.*[2] discovered the superconducting cuprate $Nd_{2-x}Ce_xCuO_{4-\delta}$, crystallizing in the T' structure and exhibiting superconductivity in a narrow composition range $0.14<x<0.18$ with a maximum critical temperature of $T_{c0}=24$ K. Since trivalent Nd is substituted with tetravalent Ce, the carriers in these compounds were expected to be electrons,[2–4] in contrast to high-T_c-cuprate superconductors, where the charge carriers are holes.[5]

Early experiments confirmed the presence of electrons as mobile charge carriers.[2] Further detailed investigations, however, complicated the issue of carrier sign: the sign of the Hall coefficient, R_H, was found to depend on x with $R_H<0$ in the range of superconducting specimens.[3,4] Other experiments yield $R_H>0$ for $x\approx 0.15$ or a sign changing with temperature.[6] Moreover, composition and temperature-dependent results for the sign of R_H were observed.[7,8] Positive thermopowers were reported[9] in conflict with the usually negative R_H for specimens with compositions in the superconducting range and high critical temperatures.[3,4,7–10]

Since the sign of the normal-state Hall coefficient and, thus the type of charge carriers is still an unresolved problem, we investigated the Hall effect and magnetoresistance of epitaxial $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ thin films. All data shown have been obtained for the same film, with optimized superconducting properties. Interpreting both quantities consistently in the same model leads to a possible explanation of the charge-carrier-type problem.

II. EXPERIMENTAL TECHNIQUES

Hollow cathode magnetron sputtering was used to grow 1200 Å thick high quality, c-axis oriented epitaxial $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ films (c-axis length 12.08 Å) on $SrTiO_3$ substrates. A 110 μm wide two Hall bars geometry[11,12] was patterned by photolithography and Ar ion beam etching. An ac lock-in method at 133 Hz with a current, I_{xx}, of 20 μA flowing parallel to the CuO_2 planes was used to measure the longitudinal voltage, V_{xx}, and the transverse voltage, V_{xy}, in a magnetic field B parallel to the c axis. The sample was mounted in an Oxford variable temperature cryostat. The temperature was detected by a carbon glass resistance thermometer. The temperature was stabilized with a precision of about 50 mK, using a capacitive sensor.

III. MAGNETOTRANSPORT

Since R_H in $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ is temperature dependent and also a magnetoresistance effect is observed, a free-carrier model is insufficient for the interpretation of our data. The simplest model beyond the one-carrier free-electron approximation is a two-band model, considering the case of a current carried by both electrons (concentration n_e, mobility μ_e, and conductivity σ_e) and holes (n_p,μ_p, and σ_p). The Hall coefficient within a two-band model is given by[13,14]

$$R_H=\frac{n_p\mu_p^2-n_e\mu_e^2}{e(n_p\mu_p+n_e\mu_e)^2}\ , \tag{1}$$

where $e>0$ is the elementary charge.

The magnetoresistance effect on the resistivity, ρ_{xx}, also can be calculated in a two-band model,[14] yielding in our experimental situation

$$\frac{\rho_{xx}(B,T)-\rho_{xx}(0,T)}{\rho_{xx}(0,T)}=\frac{\sigma_p\sigma_e(\mu_p+\mu_e)^2}{(\sigma_p+\sigma_e)^2}B^2\ . \tag{2}$$

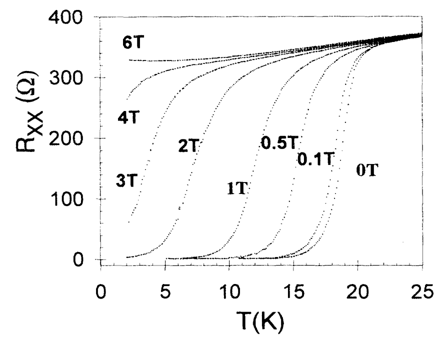

FIG. 1. Resistance, R_{xx}, vs T for different B fields.

IV. RESULTS

The transition in Fig. 1 is broadened. The zero-resistance point as well as the 90% point shift to lower temperatures for increasing B. The shape of the curves remains unaltered. Note that the 2 T curve does not reach the zero-resistance state for 2 K. There remains a resistance of 7 Ω. With increasing magnetic field the slope of the linear portion of the curves decreases slightly. The upper critical field extrapolated to zero temperature yields $B_{c2}(0)=6.3$ T, in agreement with the literature.[6]

The mixed-state Hall resistance in Fig. 2 shows a sign reversal in the lower part of the characteristics, before it approaches zero in the superconducting state. In the normal state, the Hall voltage increases with increasing magnetic field.

The normal-state Hall coefficient (Fig. 3) is negative at room temperature and decreases linearly with decreasing temperature down to 120 K. Below 120 K there is a strong increase of R_H. At 55 K the Hall coefficient shows a sign change from negative to positive. At 2 K a positive value of more than 1.25×10^{-9} m^3/C is reached, about five times larger than the absolute value at room temperature.

The longitudinal resistivity, ρ_{xx}, in zero magnetic field in Fig. 4 does not vary linearly with temperature, as usually found for high-temperature superconductors.[5] Instead, a T^2 dependence is observed.

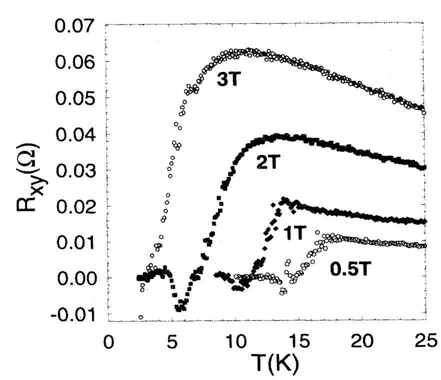

FIG. 2. Hall resistance, R_{xy}, vs T for different magnetic fields. The 2 T curve has a double sign reversal in the mixed state.

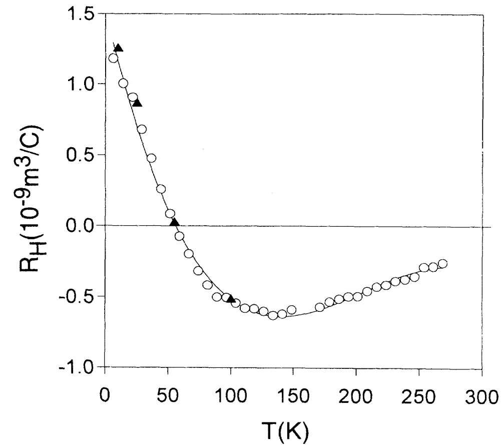

FIG. 3. Hall coefficient, R_H, vs temperature, T, for a magnetic field of $B=10$ T. Circles result from temperature scans at fixed B, and the filled triangles are from B-field scans at constant temperatures.

The magnetoresistance is positive in the temperature range, where the steep increase of the Hall coefficient, including its sign change is observed, i.e., the resistance R_{xx} increases with increasing magnetic field (Fig. 5). The magnitude of the magnetoresistance is of the order of 1%. For constant field the magnetoresistance increases with decreasing temperature.

V. DISCUSSION

The sign change of the normal-state Hall coefficient can be understood within the two-band model discussed in Sec. III, if we assume that there are two groups of charge carriers in our specimen acting independently of each other, so that their mobilities show a different temperature dependence. According to Eq. (1) the sign of R_H is then given by the difference $n_p\mu_p^2-n_e\mu_e^2$. The Hall coefficient is negative if the second term dominates,

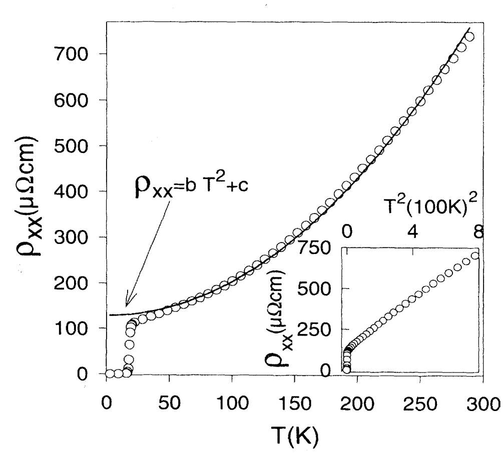

FIG. 4. In-plane resistivity $\rho_{xx}(T)$ in zero magnetic field vs T. The line is the best fit of $\rho_{xx}(T)=bT^2+c$ to the experimental data between 25 K and room temperature, yielding $b=7.55n\Omega\,\text{cm}\,\text{K}^{-2}$ and $c=128\ \mu\Omega\,\text{cm}$. The inset shows ρ_{xx} vs T^2 to demonstrate the quadratic temperature dependence.

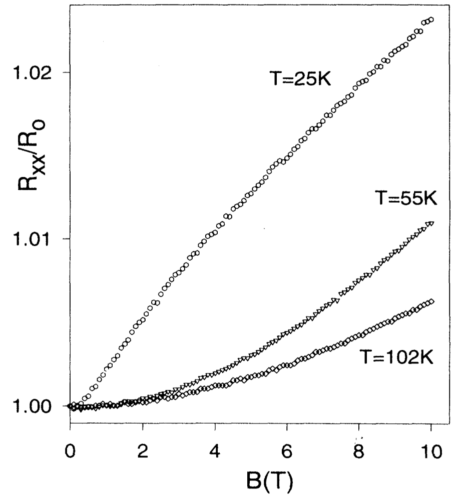

FIG. 5. Magnetoresistance, $R_{xx}(B,T)$, normalized by $R_0=R_{xx}(0,T)$ for several temperatures, T.

whereas R_H is positive if the first term dominates. If the two terms are equal in magnitude R_H becomes zero.

This interpretation is established by the behavior of the magnetoresistance. The curves in Fig. 5 can be fitted to $R_{xx}/R_0=a(T)B^2+1$ with $a(102\ \mathrm{K})=5\times10^{-4}\ \mathrm{T}^{-2}$ and $a(55\ \mathrm{K})=1.2\times10^{-4}\ \mathrm{T}^{-2}$. Since R_{xx}/R_0-1 is equal to the left-hand side of Eq. (2), the magnetoresistance for 102 and 55 K is in agreement with the two-band model. The deviation of the data at 25 K from the quadratic behavior is probably caused by superconducting fluctuations.[15]

The T^2 dependence of the zero-field resistivity shown in Fig. 4 is typical for electron-electron scattering in a three-dimensional metal. For a two-dimensional (2D) metal the T^2 law is modified by a logarithmic correction.[16] Tsuei *et al.*[17] who investigated the zero-field resistivity of $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ films, also, in a first step, fitted a T^2 law to their data, as we did in Fig. 4. They found, however, that their experimental results are better described by

$$\rho_{xx}(0,T)=\rho_{xx}(0,0)+K_{xx}(T/T_F)^2\ln(T_F/T)\ . \qquad (3)$$

Their conclusion was that $\rho_{xx}(0,T)$ is generated by electron-electron scattering and that the good fit to the logarithmically corrected T^2 law expresses the quasi-2D nature of the conductivity in their specimens. Our data for $\rho_{xx}(0,T)$ in Fig. 4 in the temperature range from 25 K to room temperature are exactly given by Eq. (3), with $\rho_{xx}(0,0)=112\ \mu\Omega\,\mathrm{cm}$, $K_{xx}=1.4\ \Omega\,\mathrm{cm}$, and the Fermi temperature $T_F=2.9\times10^4$ K as fitting parameters.

Usually, the contribution of electron-electron scattering is small and masked by electron-phonon scattering, except at very low temperatures. There are, however, special situations where the electron-electron scattering is strongly enhanced, such as "*sd* scattering" (Ref. 16) or "interpocket electron-hole scattering" (Refs. 16,18), yielding a T^2 dependence of the resistivity persisting at high temperatures. Up to now, however, no electron pockets were found experimentally at the Fermi surface, which consists of a roughly circular hole pocket.[19]

Finally, we discuss the mixed-state Hall effect: according to Josephson[20] the electric field generated by the flux motion in a type-II superconductor is given by $\mathbf{E}=-\mathbf{v}\times\mathbf{B}$, where $\mathbf{v}$ is the velocity of the flux-line structure and $\mathbf{B}$ the magnetic flux density. The Lorentz force generated by the transport current density, $\mathbf{j}_{xx}$, drives the vortex. If it would act alone, the flux-tube velocity would only have a component v_y, generating a longitudinal voltage, V_{xx}. Due to additional forces[21] the vortex motion deviates from the $\hat{\mathbf{y}}$ direction by a "Hall angle," Θ_H, with $\tan\Theta_H=v_x/v_y=E_{xy}/E_{xx}$. The standard models for the vortex motion in type-II superconductors by Bardeen and Stephen (BS) (Ref. 22) and Nozières and Vinen[23] are not able to describe the sign change of the Hall resistance in Fig. 2.

The sign change can be obtained by a two-band model, including both electrons and holes as carriers in the BS model and assuming a change of the carrier densities or mobilities on going from the mixed to the normal state.[24,25] Recently, Hirsch and Marsiglio (HM) (Ref. 25) pointed out that in high-temperature superconductors the direct overlap of oxygen orbitals leads to a hole band, while the Cu $d_{x^2-y^2}$-Op_σ orbitals yield an electron band, with a larger and a smaller superconducting energy gap, respectively. In the HM model electrons in the vortex core become "normal" more readily than the holes, at the transition to the normal state, yielding a negative Hall effect, switching to a positive Hall effect at higher temperatures as the holes become normal, too.

The effect occurs in many materials. It was observed in the low-temperature superconductors Nb and V (Ref. 26) as well as in many high-temperature superconductors, such as $YBa_2Cu_3O_{7-\delta}$,[27,28] $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$,[27] $Tl_2Ba_2Ca_2Cu_3O_{10}$,[29] and $Bi_2Sr_2Ca_1Cu_2O_{8+x}$.[12] To explain the effect in the framework of a two-band model, two separate bands must be present in these materials. On the other hand, the effect may be a general consequence of the vortex dynamics. At the moment no appropriate model is available considering this case.[27]

In conclusion, the normal-state Hall effect of $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ films exhibits a sign change. While a negative Hall effect is obtained at higher temperatures, the effect is positive at low temperatures. This observation can be interpreted within a two-band model considering two types of charge carriers, namely electrons and holes. The same model well describes the magnetoresistance observed, which increases with the quadrate of the magnetic flux density. Moreover, to understand the persistence of the quadratic temperature dependence of the zero-field resistivity up to room temperature, electron-hole scattering is invoked. The mixed-state Hall effect shows a sign anomaly, indicating that the ccmponent of the vortex velocity parallel to the transport current changes its sign.

ACKNOWLEDGMENTS

The authors would like to thank R. P. Huebener and G. Yu. Logvenov for stimulating discussions and U. Bete for technical assistance. This work was supported by the Bayerische Forschungsstiftung FORSUPRA.

[1]Y. Tokura *et al.*, Nature **337**, 345 (1989).
[2]H. Takagi *et al.*, Phys. Rev. Lett. **62**, 1197 (1989).
[3]S. Uchida *et al.*, Physica C **162-164**, 1677 (1989).
[4]H. Takagi *et al.*, Physica C **162-164**, 1001 (1989).
[5]N. P. Ong, in *Physical Properties of High Temperature Superconductors II,* edited by D. M. Ginsberg (World Scientific, Singapore, 1990).
[6]Z. Z. Wang *et al.*, Phys. Rev. B **43**, 3020 (1991).
[7]S. Kubo and M. Suzuki, Physica C **185-189**, 1251 (1991).
[8]S. J. Hagen *et al.*, Phys. Rev. B **43**, 13 606 (1991).
[9]X.-Q. Xu *et al.*, Phys. Rev. B **45**, 7356 (1992).
[10]W. Jiang *et al.*, Phys. Rev. B **47**, 8151 (1993).
[11]L. Alff *et al.*, Physica C **200**, 277 (1992).
[12]H.-C. Ri *et al.*, Phys. Rev. B **50**, 3312 (1994).
[13]C. M. Hurd, *The Hall Effect in Metals and Alloys* (Plenum, New York, 1972), Chap. 2.3.
[14]J. M. Ziman, *Principles of the Theory of Solids* (Cambridge University Press, Cambridge, England, 1989).
[15]Y. Yamasaki *et al.*, Physica C **235-240**, 1405 (1994).
[16]M. Kaveh and N. Wiser, Adv. Phys. **33**, 257 (1984).
[17]C. C. Tsuei, A. Gupta, and G. Koren, Physica C **161**, 415 (1989).
[18]P. F. Maldague and C. A. Kukkonen, Phys. Rev. B **19**, 6172 (1979).
[19]D. M. King *et al.*, Phys. Rev. Lett. **70**, 3159 (1993); R. O. Anderson *et al.*, *ibid.* **70**, 3163 (1993); S. Massidda *et al.*, Physica C **157**, 571 (1989).
[20]B. D. Josephson, Phys. Lett. **16**, 242 (1965).
[21]R. P. Huebener, in *Magnetic Flux Structures in Superconductors,* edited by P. Fulde, Springer Series in Solid State Sciences Vol. 6 (Springer, Berlin, 1979).
[22]J. Bardeen and M. J. Stephen, Phys. Rev. **140**, A1197 (1965).
[23]P. Nozières and W. F. Vinen, Philos. Mag. **14**, 667 (1966).
[24]L. C. Ho, Can. J. Phys. **48**, 1939 (1970).
[25]J. E. Hirsch and F. Marsiglio, Phys. Rev. B **43**, 424 (1991).
[26]K. Noto *et al.*, Solid State Commun. **18**, 1081 (1976).
[27]S. J. Hagen *et al.*, Phys. Rev. B **47**, 1064 (1993).
[28]J. M. Harris *et al.*, Phys. Rev. Lett. **71**, 1455 (1993).
[29]R. C. Budhani *et al.*, Phys. Rev. Lett. **71**, 621 (1993).