Universal Hall mobility in c-axis-oriented $Y_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ thin films

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Transport properties of c-axis-oriented $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films with a wide range of $x=0$–0.5 and $\delta=0.08$–1.0 have been systematically investigated. Hall mobility μ_H in all the films satisfies the relation of $\mu_H^{-1}=\alpha T^2+C$ (α and C are constants). By changing x and δ, we have successfully excluded the contribution of Cu-O chains in μ_H^{-1}, and shown that α is essentially independent of doping level and disorder. Moreover, the doping-independent α is universal in various high-T_c superconducting cuprates, and this indicates that μ_H^{-1} is more fundamental than R_{ab}^H. [S0163-1829(96)02846-9]

In recent solid-state physics, it is one of central issues whether the electronic properties in high-T_c superconducting cuprates (HTSC's) can be understood within the Fermi-liquid (FL) theory or not. Among their anomalous properties, normal-state charge transport such as the in-plane resistivity ρ_{ab} and the in-plane Hall coefficient R_{ab}^H are most strongly against the FL picture,[1] and exhibit complicated dependence on temperature,[2] carrier doping,[3–5] and disorder.[6]

An important finding on the understanding of ρ_{ab} and R_{ab}^H is that the cotangent of the Hall angle, $\cot\theta_H \equiv \rho_{ab}/HR_{ab}^H=\mu_H^{-1}/H$ is expressed as $\cot\theta_H=\alpha T^2+C$ (α and C are constants), which is valid in all the p-type HTSC's with various carrier[3,8–10] and impurity[6,9,11] concentrations. This naturally indicates that $\cot\theta_H$ (or μ_H) is more fundamental than R_{ab}^H, and thus it will be a clue for selecting HTSC theories to make clear which is fundamental. However, there is no consensus on $\cot\theta_H$ or μ_H. For example, the doping dependence of $\cot\theta_H$ in $YBa_2Cu_3O_{7-\delta}$ (YBCO) is controversial,[8–10] which might arise from the conducting Cu-O chains.[4,12]

We recently reported the transport properties of c-axis-oriented $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ ($x\leqslant 0.5$) thin films.[13] The heavy Ca doping level up to 0.5 makes it possible to dope carriers into YBCO without filling the oxygen deficiency, and thus $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films are more effective than untwinned YBCO crystals.[14] In this paper, ρ_{ab}, R_{ab}^H, and $\cot\theta_H$ of $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films have been systematically investigated. The Cu-O chain contributions have been successfully eliminated from $\cot\theta_H$, and it has been revealed that α is independent of the carrier-doping level and the disorder. Moreover, as comparing α among the HTSC's, the T-dependent part of μ_H^{-1} is found to be independent of the species of HTSC's, which strongly suggests that μ_H^{-1} is more fundamental than R_{ab}^H.

$Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films with a typical thickness of 2000 Å were deposited onto $SrTiO_3$ single-crystal substrates by RF thermal plasma deposition technique.[13] The as-grown samples were post-annealed under different thermal conditions to control the oxygen content. X-ray diffraction and transmission electron microscopy (TEM) showed that c-axis-oriented $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ was grown without any impurity phases. The Raman spectroscopy measurement revealed that the doped Ca mainly occupied the Y site. dc resistivity was measured using a standard four-probe method and Hall coefficient was measured in a magnetic field of 8 T parallel to the c axis. In order to eliminate the zero-field signal due to the misalignment of voltage contacts, we rotated the samples by 180° in a static field. The oxygen deficiency δ was estimated by the following procedure. First the hole concentration per Cu, p, was evaluated from the value of T_c using the previously reported parabolic curve $T_c/T_{c,\max}=1-82.6(p-0.16)^2$.[15] Next, δ was estimated using the relationship $p=p(\delta)$ obtained from the measurements of T_c and the thermoelectric power, and the calculation of bond-valence sum for $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ by Tallon *et al.*[16]

Figures 1(a) and 1(b) show the temperature dependence of ρ_{ab} and R_{ab}^H for $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ ($x=0$ and 0.5) thin films, respectively. First of all, we emphasize that the data

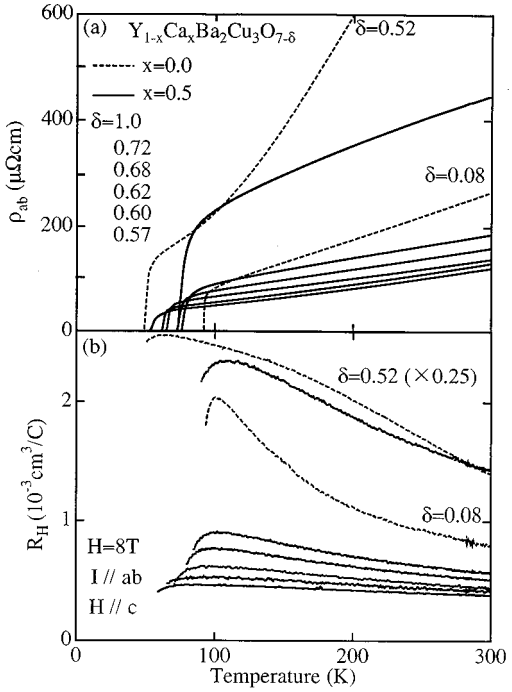

FIG. 1. Temperature dependence of (a) in-plane resistivity ρ_{ab} and (b) Hall coefficient R_{ab}^H, for $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films with various δ. The broken and solid curves are for $x=0$ and $x=0.5$, respectively.

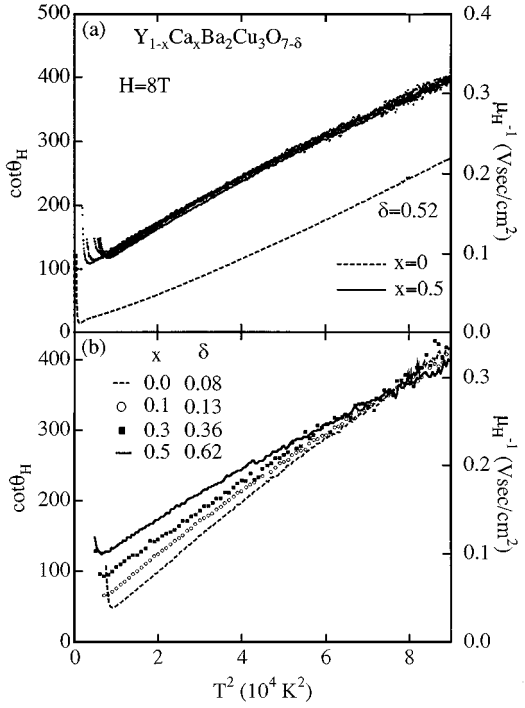

FIG. 2. T^2 dependence of $\cot\theta_H$ ($\equiv\rho_{ab}/HR_{ab}^H$) in 8 T for $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films. (a) for $x=0$, 0.5, and $\delta\geqslant 0.5$; (b) for $0\leqslant x\leqslant 0.5$ and $\delta<0.5$. The broken curves, open circles, closed squares, and solid curves correspond to $x=0$, 0.1, 0.3, and 0.5, respectively. Note that the right axis is the inverse of Hall mobilities.

for $x=0$ shown by the dotted curves are essentially identical to those for high-quality single crystals.[4] For example, the $YBa_2Cu_3O_{6.92}$ film shows the T-linear resistivity with a small residual resistivity, and shows a superconducting transition at 92 K with a reasonably sharp transition width of 1 K. These results well guarantee the quality of our samples, and consequently we think that the data for $x=0.5$ films reflect the intrinsic nature of $Y_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$. Moreover, in Ref. 13, it has been clarified that the crystallinity of the $x=0.5$ film is as high as that of $x=0$ film from TEM measurement. In the $x=0.5$ films, the doping level is varied from the under- to overdoped regions by changing δ from 1.0 down to 0.57. With decreasing δ, ρ_{ab}, and R_{ab}^H for $x=0.5$ shown by the solid curves in Fig. 1 monotonically decrease, indicating that the carriers are continuously doped. In addition, the T_c of the $x=0.5$ films takes a maximum near $\delta=0.72$ and reduces with decreasing δ. We emphasize that the optimally doped $x=0.5$ film ($\delta\sim 0.72$) has ρ_{ab} as low as $YBCO_7$, from which the mean-free path is calculated to be 21 Å at 300 K.[17] This indicates 50% Ca substitution does not disturb the in-plane conduction seriously.

The temperature dependence of $\cot\theta_H$ calculated from ρ_{ab} and R_{ab}^H in Fig. 1 is plotted as a function of T^2 in Fig. 2, where $\cot\theta_H$ in all the samples roughly satisfies the relation of $\cot\theta_H=\alpha T^2+C$. Note that the data for $\delta\geqslant 0.5$ are shown in Fig. 2(a), while the data for $\delta<0.5$ are in Fig. 2(b) with the data for $x=0.1$ and $x=0.3$ newly added. Most unexpectedly, all the $x=0.5$ samples show the identical $\cot\theta_H-T^2$ curves, i.e., both α and C are independent of δ. The δ-independent C means that the oxygen depletion does not introduce disorder into the CuO_2 plane, while the δ-independent α indicates that

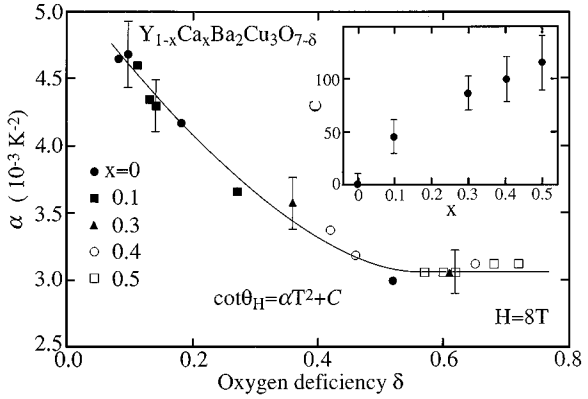

FIG. 3. The slope α of $\cot\theta_H=\alpha T^2+C$ curve for $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ ($0\leqslant x\leqslant 0.5$) thin films as a function of δ. Closed circles, closed squares, closed triangles, open circles, and open squares are for $x=0$, 0.1, 0.3, 0.4, and 0.5, respectively. The solid curve is the guide for the eyes. Inset shows the zero-temperature-intercept C as a function of x.

the carrier doping hardly affects α in $Y_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$. We further note that α for $Y_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-\delta}$ is almost equal to that for $YBa_2Cu_3O_{6.48}$. On the contrary, as shown in Fig. 2(b), the Ca substitution causes a decrease in α accompanied by an increase in C.

To see the doping dependence of α and C more clearly, we plotted α as a function of δ, and C as a function of x in Fig. 3. The Ca substitution introduces disorder into the CuO_2 plane, and thus C monotonically increases with x, whereas α is independent of x and increases with decreasing δ only for $\delta<0.5$. We think that the large α in the $\delta\sim 0$ samples arises from the conducting Cu-O chains. Reflecting the one-dimensional nature, the Cu-O chains are expected to give no Hall voltage. In addition, in real experimental situations, Hall bars electronically short the CuO_2 plane to the Cu-O chain, which force us to reduce the Hall voltage in the plane to that in the chain.[18] The fact that $YBa_2Cu_4O_8$ with conducting Cu-O double chains shows unusually small R_{ab}^H (Ref. 19) evidences our interpretation. Accordingly, the δ-independent α for $\delta\geqslant 0.5$, where the Cu-O chains are no longer conductive, can be regarded as the intrinsic value to the CuO_2 plane in YBCO.

Since we have shown that the slope α inherent in the CuO_2 plane in YBCO is independent of carrier concentration and disorder, let us now compare μ_H^{-1} for YBCO with that for other HTSC's. We have selected the data to be compared on the basis of following criteria. (i) μ_H^{-1} should be measured using high-quality single crystals, because resistivity is usually one order of magnitude larger in polycrystals than in single crystals. (ii) Overdoped $La_{2-x}Sr_xCuO_4$ should be excluded as an exception. La-Sr-Cu-O is the only p-type cuprate whose R_{ab}^H changes its sign in the overdoped region, and thus μ_H^{-1} diverges when $R_{ab}^H\sim 0$.[20,21]

Figure 4 shows thus selected μ_H^{-1} as a function of T^2 for various HTSC's (Refs. 3, 5, 22, and 23), where the T-dependent part in μ_H^{-1} suprisingly coincides among all the data. It is our claim in the present paper that *the T-dependent part in μ_H^{-1} is universal regardless of carrier concentration, disorder, and class of HTSC's.*

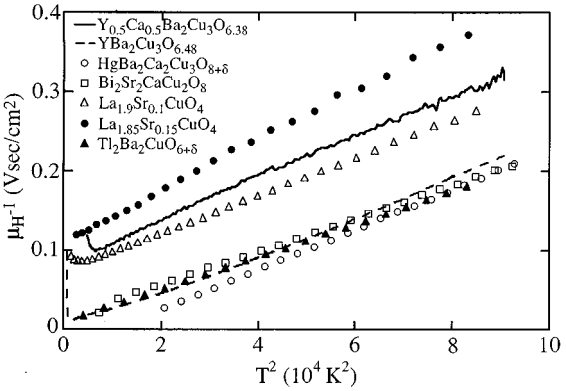

FIG. 4. The comparison of the inverse Hall mobility μ_H^{-1} as a function of T^2 for various HTSC's. The solid curve and broken curve are for the present $Y_{0.5}Ca_{0.5}Ba_2Cu_3O_{6.38}$ and $YBa_2Cu_3O_{6.48}$ thin films, respectively. The open triangles and closed circles are for the data of $La_{2-x}Sr_xCuO_4$ with $x=0.1$ and 0.15, respectively (Ref. 5). The open squares, closed triangles, and open circles are for the data of $Bi_2Sr_2CaCu_2O_8$ (Ref. 22), $TlBa_2CuO_{6+\delta}$ (Ref. 3), and $HgBa_2Ca_2Cu_3O_{8+\delta}$ (Ref. 23), respectively.

Here we will consider the physical meaning of the universal T dependence in μ_H^{-1} qualitatively. We can point out many facts implying that μ_H^{-1} is related to fermion-fermion scattering. First, the T-dependent part in μ_H^{-1} is roughly proportional to T^2, which reminds us of electron-electron scattering. Second, the doping independence in μ_H^{-1} suggests that μ_H^{-1} is associated with $1-p$, that is, the large Fermi surface predicted by the band calculation. Third, the remarkable difference in μ_H^{-1} between overdoped La-Sr-Cu-O and other HTSC's is attributed to the different shapes of the Fermi surface; the band theory has predicted that the Fermi surface in La-Sr-Cu-O goes from open to close with carrier doping,[24] whereas that in other HTSC's remains open.[25–27] Taking these facts into account, we suggest that the universal μ_H^{-1} is a direct probe for the uncorrelated Fermi surface. Note that, even in non-Fermi liquids, the shape of the Fermi surface will more or less influence the electronic properties. As long as elementary excitations derived from electrons are fermions, any properties related to the Fermi-Dirac statistics will remain unchanged. For example, the Fermi surface of electrons survives as the Fermi surface of spinons in the resonating-valence-bond (RVB) theory, as suggested by Tanamoto, Kohno, and Fukuyama.[28] From the viewpoint of the RVB theory, μ_H^{-1} is related to spinon-spinon scattering proposed by Anderson,[7] which is expected to coincide with the electron-electron scattering in the overdoped HTSC's.

Finally we will comment on the theories[3,7,22,29,30,31] explaining the T^2 dependence of μ_H^{-1} briefly. Our results clearly indicate that μ_H^{-1} is more fundamental than ρ_{ab} or R_{ab}^H, and hence any theories giving explicit expressions to ρ_{ab} and R_{ab}^H separately are unlikely to explain the universal μ_H^{-1}. It is very hard to believe that ρ_{ab} or R_{ab}^H, which change with doping and disorder in a complicated way, accidentally keep ρ_{ab}/R_{ab}^H unchanged. After such theories are ruled out, there remain only two possibilities, to our knowledge: one is proposed by Anderson[7] and the other is suggested independently by Kendziora *et al.*[22] and Carrington *et al.*[29] on the basis of FL. Both explanations assume two kinds of scattering times. μ_H^{-1} is attributed to spinon-spinon scattering in the former, while it is assigned to the scattering at the corner of the rounded-square-shaped Fermi surface in the latter. Although the present data are still insufficient to choose either of the two as a correct answer, we expect that the T dependence of ρ_{ab} is difficult to explain within a FL picture. For example, ρ_{ab} for $x=0.5$ and $\delta=1.0$ is slightly sublinear as a function of T, and the T-dependent part in ρ_{ab} for $x=0.5$ and $\delta\leqslant 0.72$ is not proportional to T^2. In other words, ρ_{ab} for $x=0.5$ films does not show a typical T dependence for HTSC's, possibly owing to the highly disordered CuO_2 plane induced by the 50 at. % substitution of Ca for Y. These results suggest that conventional Matthiessen's rule expected in a FL picture is seriously violated. For the elucidation of the physical meaning of the universal μ_H^{-1} and the effect of disorder on ρ_{ab}, further quantitative studies are necessary both experimentally and theoretically.

In summary, the transport properties of $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ thin films with a wide range of $x=0.0$–0.5 and $\delta=0.08$–1.0 have been systematically studied. For all the samples, the T-dependent part of μ_H^{-1} is roughly proportional to T^2. By changing x and δ, we have successfully extracted the inherent element of the CuO_2 planes in μ_H^{-1}, and shown that the T-dependent part of μ_H^{-1} is essentially independent of doping level and disorder. We have further found that the T-dependent part in μ_H^{-1} is universal in various HTSC's, which strongly suggests that μ_H^{-1} is more fundamental than R_{ab}^H, and the existence of two kinds of scattering times.

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[18] This is the same situation as Hall bars too close to a current pad giving a smaller voltage than the intrinsic value; In a two-band model with a two-dimensional plane band and a one-dimensional chain band, assuming $\rho_{\rm pl}\approx\rho_{\rm ch}$, one can obtain $\cot\theta_{\rm tw}=4/3\cot\theta_{\rm pl}$, where $\cot\theta_{\rm tw}$ is the Hall angle for a twinned crystal and $\cot\theta_{\rm pl}$ is the Hall angle inherent in the CuO_2 plane. Thus, the increase of α with decreasing δ is also explainable with the two-band model. In our real observation, however, the increase of α is larger than the calculated one.

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