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Influence of the oxygen content on the normal-state Hall angle in $YBa_2Cu_3O_{x_n}$ films

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We report on measurements of in-plane resistivity $\rho_{ab}(T)$ and Hall coefficient $R_H(T)$ in c-axis-oriented oxygen-deficient $YBa_2Cu_3O_{x_n}$ thin films. Independent of the changes induced in $R_H(T)$ and $\rho_{ab}(T)$ by the oxygen depletion, a quadratic temperature dependence is observed for the Hall angle θ_H. The slope of $\cot\theta_H$ versus T^2 decreases as the oxygen content x_n is reduced and saturates in the $T_c(x_n)=60$ K plateau region. The results are discussed in the framework of the two-dimensional Luttinger liquid theory as proposed by Anderson.

Experimentally, the temperature-dependent Hall coefficient $R_H(T)$ observed in high-T_c superconductors seems to be a universal characteristic property of these materials.[1] In samples with an optimum hole doping level (i.e., a maximum T_c), the temperature dependence of the Hall coefficient, $R_H \propto 1/T$, coincides with a linear temperature dependence of the in-plane resistivity, i.e., $\rho_{ab}(T)=\beta T$, with $\rho_{ab}(0)=0$.[2] In most cases, however, the temperature dependence of R_H is not well defined. Theoretically, it became evident that the temperature dependence of R_H does not correspond to the one-band Fermi-liquid description of R_H. Several attempts have, however, been made to explain the unusual normal-state transport properties of high-T_c materials within the Fermi-liquid theory.[3] Models including two or more energy bands,[4] narrow metallic impurity bands,[5] magnetic skew scattering,[6] and carrier concentration dependent bandwidths[7] have been proposed.

A possible breakthrough in the Hall effect "puzzle" was initiated by the experiments of Chien, Wang, and Ong[8] in $YBa_2Cu_{3-x}Zn_xO_7$ single crystals. By computing the Hall angle θ_H as a function of temperature, a universal quadratic temperature dependence of $\cot\theta_H$ was observed for various Zn concentrations:

$$\cot\theta_H=\frac{\rho_{ab}}{R_HB}=\alpha T^2+C \ , \qquad (1)$$

where B is the applied magnetic field, α is a constant ($\alpha=5.11\times10^{-3}$ K^{-2} at 8 T) for all Zn concentrations, x, and C is a linear function of x. More recently, an identical behavior of $\cot\theta_H$ was observed in $Y_{1-x}Pr_xBa_2Cu_3O_7$ single crystals for $x\leq0.55$,[9] in $La_{1.85}Sr_{0.15}Cu_{1-x}A_xO_4$ ceramic samples with A =Fe,Co,Ni,Zn,Ga, for a wide range of impurity concentrations,[10] and in overdoped Tl cuprates.[11] These results strongly suggest that the quadratic temperature dependence of the Hall angle is closely related to the mechanism responsible for the anomalous normal-state properties of high-T_c superconductors.

The experiments of Chien, Wang, and Ong[8] were explained by Anderson in the framework of the two-dimensional Luttinger liquid theory.[12] The basic mechanism in this model is related to the existence in the normal state of two types of quasiparticle excitations with different electronic relaxation rates. On the one hand, the transport relaxation time $\tau_{tr}\propto T^{-1}$, due to the scattering between "holons" and "spinons," which determines the longitudinal conductivity σ_{xx}; on the other hand, the transverse relaxation time, $\tau_H\propto T^{-2}$, determined by scattering between spinons alone. Since spinons can also interact with magnetic impurities, a temperature-independent relaxation time can be added to τ_H using Matthiessen's rule.

More recently, other models for the T^2 temperature dependence of the Hall angle have been proposed. Kubo and Manako[11] claim that their observations in Tl-based overdoped cuprates can be explained by assuming a temperature-dependent carrier density n and only one relaxation time $\tau\propto T^{-2}$. Levin and Quader[13] propose a subband phenomenological model which also predicts the existence of two types of quasiparticles with different relaxation times and a T^2 dependence for $\cot\theta_H$. Finally, Ushio, Schimizu, and Kamimura[14] interpret the temperature dependence of the Hall angle as being entirely due to the warped shape of the Fermi surface in high-T_c materials.

Contrary to the Pr and Zn substitutions in $YBa_2Cu_3O_7$, which apparently do not alter the carrier density,[8,9] it is generally accepted that a reduction of the oxygen content leads to a corresponding decrease of n in the CuO_2 planes. We therefore performed systematic Hall coefficient $R_H(T)$ and in-plane resistivity $\rho_{ab}(T)$ measurements in oxygen-deficient $YBa_2Cu_3O_{x_n}$ (YBCO) films, in order to study the behavior of the Hall angle θ_H as a function of a varying carrier density.

Thin films of $YBa_2Cu_3O_7$ are deposited onto MgO(100) single-crystal substrates by an *in situ* 90° off-axis dc sputtering technique using a single stoichiometric (YBCO) ceramic target. Details of the thin-film preparation procedure have been published elsewhere.[15] X-ray-diffraction characterization shows that the 110-nm-thick films grow epitaxially with the c axis perpendicular to the substrate plane. The as-prepared films have a high critical temperature ($T_c\simeq89$ K, $\Delta T_c<2$ K), and critical current density ($J_c\simeq10^7$ A/cm^2 at 5 K, $J_c\simeq10^6$ A/cm^2 at

77 K). The films are patterned using classical photolithography and wet etching techniques producing a (2×10) mm^2 pattern with the necessary voltage and current contacts.

Oxygen-deficient films are made by a simple procedure reported previously.[16] The film is packed in a box of YBCO bulk material and placed inside a quartz tube. The desired oxygen concentrations are obtained by a controlled heat treatment of the film following a constant oxygen content (x_n) line in the oxygen pressure-temperature (P_{O_2}-T) phase diagram of $YBa_2Cu_3O_x$.[17] Systematic critical temperature and x-ray-diffraction experiments revealed that films with different oxygen contents ($6\leq x_n\leq 7$) can be obtained in a controlled, reproducible, and reversible way. Moreover, the oxygen-deficient films have featureless narrow resistive transitions, which points to a homogeneous oxygen distribution in the films. All the oxygen concentrations (x_n values) reported in this paper are nominal values derived from the P_{O_2}-T phase lines.

The normal and superconducting properties of the YBCO films are measured in a temperature stabilized He flow cryostat placed inside a rotating electromagnet (temperature stability $\simeq$ 100 mK; maximum field $B=1$ T). All electrical contacts are made by direct wire bonding using AlSi wires onto the film. The resistivity was measured up to room temperature using a four-probe ac technique (transport current $I<300$ μA). The Hall coefficient was measured in a magnetic field $B=0.72$ T using a standard field inversion technique and averaging over a large number of measurements at each temperature. For all the x_n values the Hall voltage is linear with field and current at room temperature. In one film ($x_n=6.7$), the linearity of the Hall voltage versus magnetic field was checked at several temperatures ($T=80$, 100, 140, and 300 K) for fields up to 12 T using a superconducting magnet. No indication of a saturation in the Hall voltage was observed.

In the present study we report on $\rho_{ab}(T)$ and $R_H(T)$ measurements performed on the same YBCO film which was successively oxygen depleted to contain (in this order) $x_n=6.85$, 6.75, 6.9, 6.7, 7.0, and 6.6 oxygen atoms per unit cell. The reproducibility of the results has been checked by repeating the measurements in two other YBCO films.

The temperature dependence of the resistivity for the YBCO film with different oxygen contents x_n is shown in Fig. 1. It is clear that the absolute value of the resistivity increases with decreasing oxygen content and that the linear temperature dependence of ρ_{ab} is drastically altered for $x_n<6.90$. This feature, together with the fact that $d\rho_{ab}/dT$ increases when x_n is reduced[18] seems to be characteristic for oxygen-deficient YBCO samples. An increase of $d\rho_{ab}/dT$ has also been observed in the $La_{2-z}Sr_zCuO_4$ system,[19] when z is reduced from its optimum value $z=0.15$. The normal to superconducting transition width stays very narrow ($\Delta T_c\leq 2$ K) for all oxygen contents (except for $x_n=6.6$ for which $\Delta T_c\simeq 5$ K), indicating a good oxygen homogeneity in the film. The critical temperature as a function of x_n decreases in a similar way as reported for bulk YBCO,[20] i.e., showing two plateaus at, respectively, 90 K and 55 K.

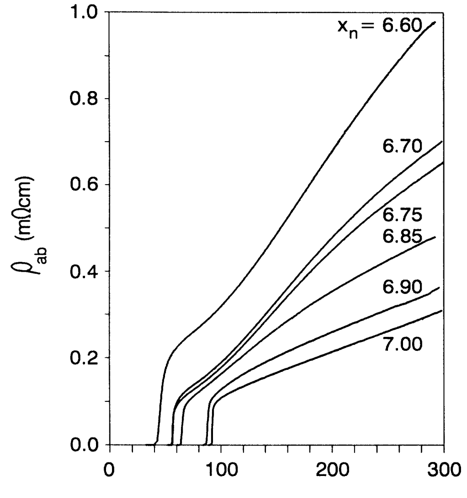

FIG. 1. Measured in-plane resistivity ρ_{ab} as a function of temperature for a YBCO film with different nominal oxygen contents x_n.

Figure 2 shows the temperature dependence of the inverse Hall coefficient $1/R_H(T)$ obtained from the same film as in Fig. 1, for different oxygen contents. It is evident that $1/R_H$ decreases with decreasing x_n over the whole temperature range and has a pronounced temperature dependence. For the higher x_n values, $1/R_H$ varies linearly with T down to a few degrees above T_c. For the lower x_n values ($x_n\leq 6.85$), the region of linearity systematically shrinks. A similar behavior of $1/R_H$ has been reported in other cuprates.[1]

Using the measured values of the resistivity and the inverse Hall coefficient, the computed value of $\cot\theta_H=\rho_{ab}/R_HB$ ($B=0.72$ T) as a function of T^2 is shown in Fig. 3. For the higher oxygen contents

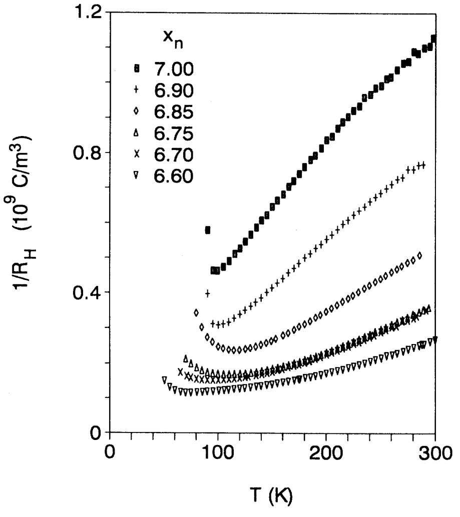

FIG. 2. Temperature dependence of the inverse Hall coefficient $1/R_H$, measured in the same YBCO film as shown in Fig. 1.

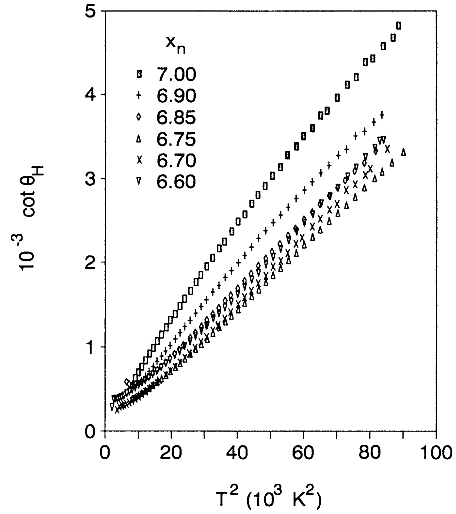

FIG. 3. Cotangent of the Hall angle θ_H at $B=0.72$ T as a function of T^2 for different oxygen contents x_n in the YBCO film.

($x_n \geq 6.9$), a linear behavior is observed between $T \simeq 100$ K and $T \simeq 240$ K. For $x_n \leq 6.85$, cotθ_H is linear in T^2 up to room temperature, but departs from linearity below 140 K. The low-temperature deviations may be due to superconducting fluctuations above T_c or weak localization effects. The origin of the deviations at high temperature, also observed in some samples by Chien, Wang, and Ong,[8] has, however, not yet been elucidated. Nevertheless, it is quite remarkable that a quadratic temperature dependence of cotθ_H is present for all x_n values, irrespective of the complicated temperature dependence of the resistivity ρ_{ab} (Fig. 1) and inverse Hall coefficient $1/R_H$ (Fig. 2). In all cases the data can be *extrapolated to zero* within the experimental error, regardless of the finite values of $1/R_H$ ($T=0$) and ρ_{ab} ($T=0$). Finally, it is also important to note that the slope α of cotθ_H versus T^2 *systematically decreases* with decreasing oxygen concentration x_n. This is clearly different from the behavior reported for Pr- or Zn-substituted YBCO crystals.[8,9]

Chien, Wang, and Ong[8] derived a relation between the bandwidth W_s of the spin carriers (spinons) and the slope α:

$$\alpha B = k_B^2 \phi_0 n / W_s^2 , \tag{2}$$

with n the two-dimensional carrier density, $\phi_0 = h/e$ the flux quantum, and k_B the Boltzmann constant. From Eq. (2) it is clear that the ratio n/W_s^2 should be temperature independent. Whether n and W_s are both constant or temperature dependent has, however, not yet been clarified.

Recently, it was observed that after Pr or Zn substitutions the value of α remained constant, suggesting that W_s remains unaffected since the carrier density does not vary for these substitutions.[8,9] On the other hand, it is well known that a reduction of the oxygen content in YBCO produces a decrease in n.[21] Assuming that T_c varies linearly with n,[22] we expect that the product $\alpha B(x_n)$ behaves in a similar way as $T_c(x_n)$ if the spinon bandwidth W_s does not depend on the carrier concentration. In order to illustrate this point we plotted $\alpha B(x_n)$ and $T_c(x_n)$, measured in the same oxygen-deficient film, in, respectively, Figs. 4(a) and 4(b). It is evident that αB not only decreases with decreasing oxygen content, but has also the tendency to saturate in the x_n region where $T_c(x_n)$ displays the 60-K plateau [Fig. 4(b)]. It should also be noted that the value $\alpha B \simeq 4 \times 10^{-2}$ T/K^2 obtained for the YBCO film with $x_n = 7$ is in excellent agreement with the values reported in Refs. 8 and 9 for fully oxidized YBCO single crystals. This is remarkable in view of the fact that the $1/R_H$ and the ρ_{ab} values are significantly different between these samples. These results strongly suggest that the product αB is indeed determined by the carrier concentration in the oxide superconductors, as predicted by Eq. (2). However, a full description requires an analysis of the variation of W_s as a function of hole doping, in the framework of Anderson's model. On the other hand, several attempts have been made to describe the normal-state properties of high-T_c materials as a function of hole concentration within a Fermi-liquid model.[3-7] The loss of the linear temperature dependence of both $\rho_{ab}(T)$ and $R_H(T)$ as x_n is reduced (Figs. 1 and 2), and the striking universal behavior of cotθ_H versus T^2 (Fig. 3) are, however, difficult to explain within these models.[4-7] For example, none of the models[4-7,13] is able to reproduce the increase in $d\rho_{ab}/dT$ (see Fig. 1 and Refs. 18 and 19) or the decrease in the slope α of cotθ_H versus T^2, when n is reduced.

In conclusion, measurements of the resistivity and Hall effect in oxygen-deficient YBCO films enabled us to address some fundamental problems related to the normal-state properties of high-T_c materials. The main observa-

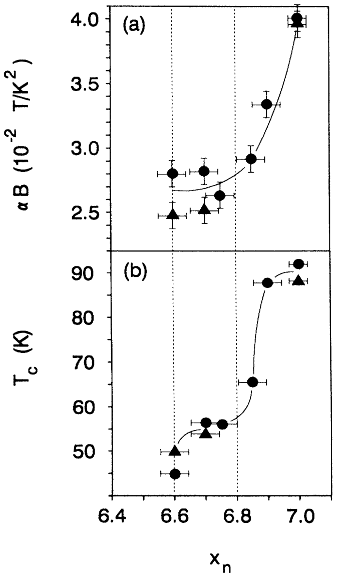

FIG. 4. Influence of the nominal oxygen content x_n on (a) the product of α (slope of cotθ_H vs T^2) and B (applied magnetic field), and (b) the critical temperature T_c. The symbols ● and ▲ correspond to two different YBCO films. The lines are a guide to the eye.

tions can be summarized as follows. Irrespective of the complicated temperature dependence of ρ_{ab} and R_H, the Hall angle θ_H shows a universal quadratic temperature dependence. A reduction of the oxygen content produces a decrease in the slope α of $\cot\theta_H$ versus T^2, in sharp contrast to the results obtained in YBCO single crystals with Pr or Zn substitutions. Finally, the dependence of αB and T_c on the oxygen concentration x_n seems to be closely related.

We recently became aware of Hall effect measurements in Co-substituted YBCO single crystals reported by Carrington *et al.* [Phys. Rev. Lett. **69**, 2855 (1992)]. They observed a quadratic temperature dependence of the Hall angle for all substitution levels, with deviations at high temperature for the low-doped samples. Moreover, similar to our findings in oxygen-deficient YBCO films, a close relation between the slope of $\cot\theta_H$ versus T^2 and T_c can be observed.

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