

Hall-effect study of bulk $\text{YBa}_2\text{Cu}_{3-x}\text{Fe}_x\text{O}_y$ ($0 \leq x \leq 0.2$, $6 < y < 7$)

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We report systematic measurements of the resistivity and of the Hall coefficient R_H of bulk $\text{YBa}_2\text{Cu}_{3-x}\text{Fe}_x\text{O}_y$ by varying both the iron doping and the oxygen concentration and for temperatures ranging from 100 to 250 K. The Hall number n_{cell} (at 100 and 200 K) decreases linearly with decreasing y for all x and exponentially upon Fe doping. The temperature dependence of n_{cell} is strongly suppressed upon removing holes from the system either by increasing the amount of iron doping or by de-oxygenation. The dependence of n_{cell} on temperature and on doping as well as the relation between $dn_{\text{cell}}/dT - T_c$ and $T_c - n_{\text{cell}}$ are also discussed. Finally, we discuss our results in the context of the $\cot\theta_H$ vs T^2 variation.

INTRODUCTION

The elucidation of the normal-state properties of the high- T_c superconductors is probably the key for the understanding of the mechanism that is responsible for the occurrence of superconductivity in the cuprates. Among the most powerful probes is certainly the Hall effect since it provides information on the carrier density and the sign of the carriers as well.

Studies on single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_y$ (1:2:3) showed that the Hall coefficient R_H is positive for $\mathbf{H} \parallel \mathbf{c}$, with an unusual $1/T$ dependence, while it is negative for $\mathbf{H} \perp \mathbf{c}$ with very little temperature dependence.¹ Since a $1/T$ dependence is also met in ceramic samples,²⁻⁴ the CuO_2 planes must dominate the Hall coefficient. The $1/T$ dependence remains still unexplained⁵ and is most pronounced for the 1:2:3 superconductor, but there is increasing evidence that R_H behaves similarly in the other cuprates as well.⁶⁻⁸

Experiments in Zn-doped 1:2:3 single crystals⁹ proved that the Hall anomaly can be better described in terms of the cotangent of the Hall angle. It was found that $\cot\theta_H = \alpha T^2 + C$. In an accompanying letter, Anderson¹⁰ in view of the fact that these systems are in the strong correlation limit, proposed an explanation within the two-dimensional Luttinger liquid framework. Recently, Carrington *et al.*¹¹ explained the unusual T dependence of R_H and the T^2 dependence of $\cot\theta_H$ by a simple model involving a two-dimensional (2D) Fermi surface with relatively large flat regions and small rounded corners (as calculated by Pickett *et al.*¹² for the CuO_2 planes of $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$). Finally, Kubo and Manako¹³ explained their data in T1-based overdoped cuprates by assuming a temperature-dependent carrier density and only one relaxation rate $1/\tau \propto T^2$. However, their model does not seem to account for the observed linear thermopower.¹⁴

Experiments in Ni- (Ref. 15), Co- (Ref. 11), and Pr- (Ref. 16) doped 1:2:3: single crystals, c -axis-oriented thin films,^{17,18} $\text{La}_{1.85}\text{Sr}_{0.15}\text{Cu}_{1-x}\text{A}_x\text{O}_4$ ceramic samples with $A = \text{Fe, Co, Ni, Zn, Ga}$,¹⁹ and overdoped T1 cuprates¹³ have proved the generality of the $\cot\theta_H$ behavior.

Regarding 1:2:3, the effect of oxygen stoichiometry and of 3d-transition metal doping (Fe, Co, Ni, Zn) on the value of R_H as well as on the temperature dependence of R_H have been widely studied.^{3,4,20-22} R_H remains positive but its temperature dependence is systematically suppressed upon doping (either by de-oxygenation or by 3d-metal doping), following the depression of the T_c . It was found that Ni or Zn doping does not alter significantly the Hall number n_{cell} (defined as $n_{\text{cell}} = V_{\text{cell}}/R_H e$, where V_{cell} is the unit cell volume) whereas oxygen depletion and Co or Fe doping affect considerably n_{cell} , suggesting that the mechanism which is responsible for the destruction of the superconducting state is different in the two cases.

Following our previous work on the influence of a controlled variation of both the iron and the oxygen content on the structural parameters and on the superconducting parameters (such as T_c and Meissner fraction) of the 1:2:3 superconductor,²³ we come now to study the effect of this simultaneous variation on the Hall number as well as on the temperature dependence of n_{cell} . Moreover, we discuss our results in terms of the $\cot\theta_H$.

EXPERIMENTAL PROCEDURES

Samples of $\text{YBa}_2\text{Cu}_{3-x}\text{Fe}_x\text{O}_y$ with $x = 0, 0.5, 0.1, 0.15$, and 0.2 were prepared with the standard solid-state reaction technique. Before the final state of the 550°C oxygenation (that is necessary in order to reach $y = 7$) each pellet was pulverized and the resulting powder was equally divided in small batches. Then, each batch was pressed in the form of a rectangular bar with typical dimensions $19.5 \times 6.5 \times 1.2$ mm. These rectangular bars were sintered under flowing O_2 at 980°C for 24 h, held under flowing O_2 at 550°C for another 24 h and they were allowed to cool to room temperature inside the furnace. Thermogravimetric analysis (TGA) showed that the oxygen content of the as-prepared samples was 6.95 ± 0.05 .

The samples with the variable oxygen content were prepared according to the following procedure. First, we

obtained the TGA traces of the as-prepared samples (when in powder form) by heating them up to 900°C in flowing Ar and with a heating rate of $4^\circ\text{C}/\text{min}$. In this way we could immediately find at which temperature a given loss of oxygen was completed. Thus, after deciding which samples we wanted to prepare (with regard to the oxygen loss), we heated each rectangular bar up to the corresponding temperature with a rate of $1^\circ\text{C}/\text{min}$ (since the oxygen kinetics are much slower in the case of the pellet) under flowing Ar in a completely programmable furnace. When the desired temperature was reached the rectangular bar was immediately cooled to room temperature and kept in the Ar flow for more than 1 h before it was removed from the furnace (to ensure that there was no oxygen uptake from the air). Since the T_c of our samples are nearly the same with those in our previous work²³ on Fe-doped 1:2:3, we are sure that their oxygen contents are the ones we deduced from the thermogravimetric analysis, within of course the experimental error of the technique.

Six electrical contacts were made using silver paste on silver pads that were deposited on the samples with dc magnetron sputtering. Hall measurements were conducted in a magnetic field of 6 T and the linearity of the Hall voltage vs B was checked for at least two temperatures in fields of 1–6 T. The measuring current was between 10 and 100 mA. The T_c of the samples were determined as the midpoint (90–10 %) of the resistive transition.

RESULTS

The identification of the Hall number n_{cell} with the actual carrier density per unit cell volume is valid only for parabolic band structure and isotropic scattering. In general, the Hall coefficient is a complicated average of the local Fermi surface curvature and it is the curvature that determines its sign and not the carrier type. Moreover, the fact that in the cuprates R_H is strongly T dependent probably makes unrealistic a simple inference of the actual carrier density from R_H . However, a lot of experimental results were described and discussed in terms of n_{cell} (which can best be thought of as an effective carrier density). In order to compare our results with previous work and to discuss certain points we will also use the Hall number convention. On the other hand, we discuss our results in terms of the cotangent of the Hall angle despite the polycrystalline nature of our samples (in sintered samples Hall measurements are greatly influenced by the c -axis resistivity and grain boundary effects) and the limited temperature range.

The $\rho(T)$ curves of the as-prepared samples are presented in Fig. 1(a). All the samples appear to have a metallic normal state. The width of the resistive transition ΔT_c increases as the Fe content increases. For the $x=0$ sample the transition width is only about 0.5 K, while for the $x=0.2$ sample $\Delta T_c \approx 8$ K.

The temperature dependence of the resistivity has been measured for all the series of samples corresponding to the various x . Representative results for the $x=0.05$ series of samples are shown in Fig. 1(b). The normal-state behavior changes from metallic to semiconducting-

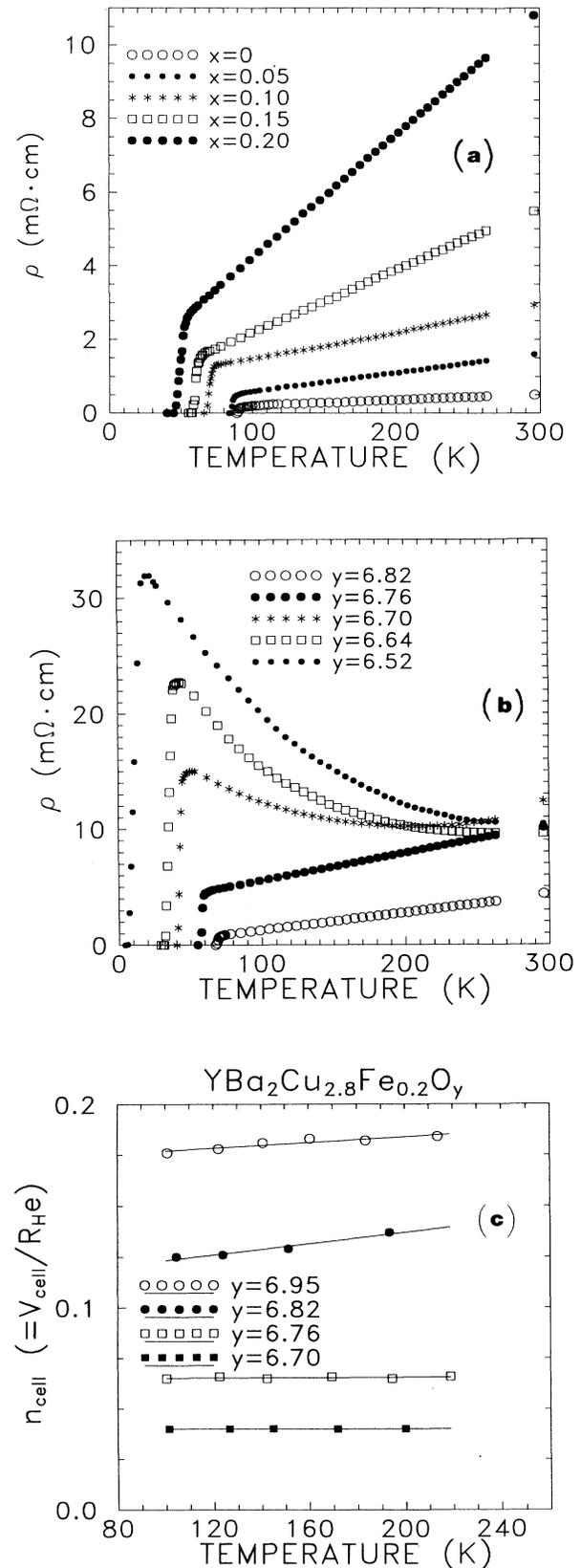


FIG. 1. The $\rho(T)$ curves for (a) the as-prepared samples and (b) the $x=0.05$ series.

like as the oxygen content decreases. For the $x=0.2$ series, all the samples (except the as-prepared sample) show a semiconductinglike normal state.

The systematic nature of the variation of the temperature dependence of the resistivity as a function of iron doping and oxygen content for all the samples (as-prepared and de-oxygenated) is convincing evidence for the reliability of the preparation procedure.

The variation of the Hall number with temperature for the as-prepared samples is shown in Fig. 2 (the error bars in all figures denote the experimental error and data points without error bars mean that the size of the centered symbol used is greater than the experimental error). It can be seen that n_{cell} shows a linear T dependence with a slope that is gradually suppressed with increasing Fe doping until it becomes negligible at $x=0.2$ and also that the implied carrier concentration decreases as more Fe enters into the structure. Substituting Fe^{3+} for Cu^{2+} is expected to lead to a decrease of the hole density but it should be noted that the oxygen content of 1:2:3 is known to increase upon Fe doping.²⁴ For the $x=0.2$ as-prepared sample n_{cell} is estimated to be 0.18 holes/cell at 100 K and it is almost insensitive to the variation of the temperature. The overall behavior resembles what has been reported for the case of Co-doped 1:2:3.²¹

For the series of samples corresponding to various x , we can see in Fig. 3 that the removal of oxygen decreases n_{cell} (as expected) and also weakens its linear T dependence.

Figure 4 shows the variation of n_{cell} with the Fe content at $T=200$ K and for various oxygen stoichiometries. This variation can be described with an exponential law for all the oxygen contents (the 100 K values also show this behavior even though the exponential decrease is much slower). The exponential decrease of n_{cell} with Fe doping becomes faster as more oxygen is removed from the structure. The n_{cell} vs x variation for the as-prepared

samples come to terms with what has been reported for Fe- and Co-doped 1:2:3.^{21,22}

Figure 5 shows the variation of n_{cell} with oxygen stoichiometry for the undoped compound as well as for various Fe dopings at 200 K. At 100 K the situation is completely analogous. The plots are linear with different slopes and intercepts. Recently, Cooper *et al.*⁴ reported that for the undoped 1:2:3 the normalized Hall number [$n_{\text{cell}}(y)/n_{\text{cell}}(y=7)$] at room temperature decreases exponentially with decreasing y . However, the absolute values of n_{cell} we measured at 100 K (and at 200 K as well) for the $x=0$ series of samples are in excellent agreement with those in Ref. 4. The plateau profile in the n_{cell} vs y variation reported by Wang *et al.*³ was not observed either in this work or in the work of Cooper *et al.*

Next, we discuss the variation of the T_c with the Hall number for the as-prepared samples. At 200 K, T_c ap-

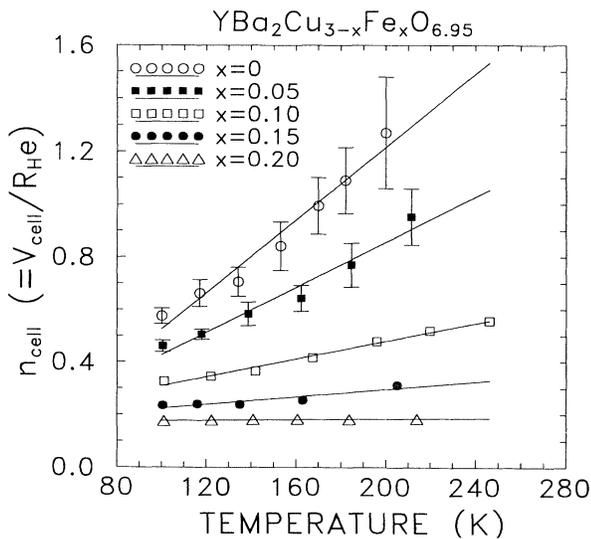


FIG. 2. The variation of n_{cell} with temperature for the as-prepared samples.

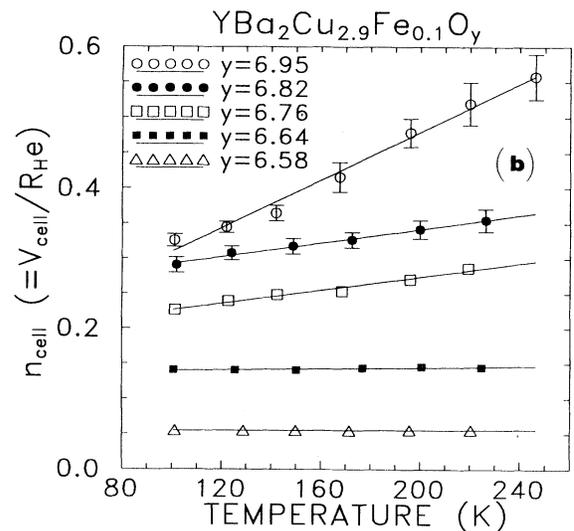
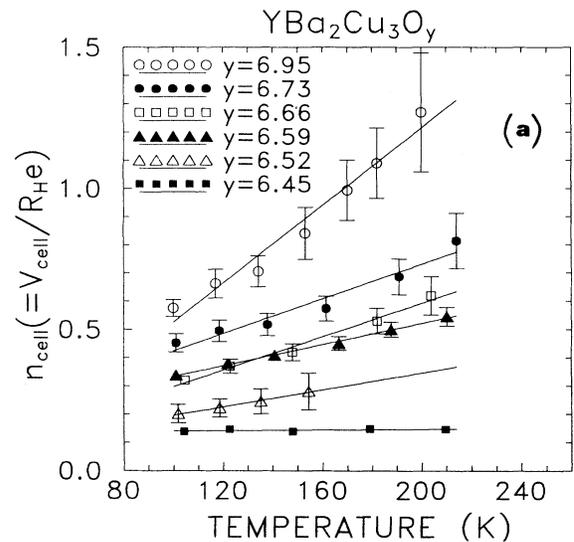


FIG. 3. The variation of n_{cell} with temperature of the various series of samples (a) $x=0$, (b) $x=0.1$, and (c) $x=0.2$.

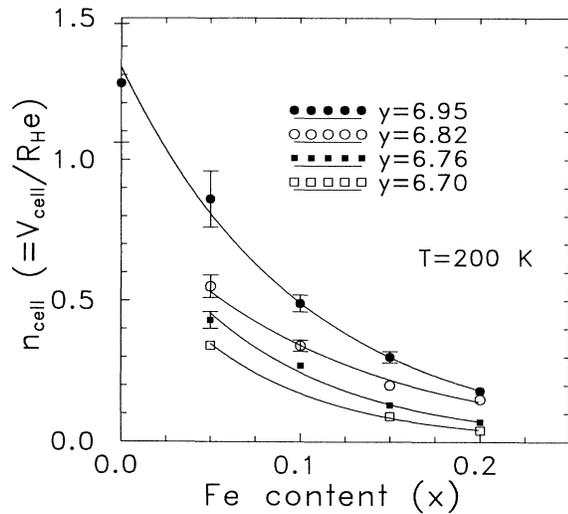


FIG. 4. The variation of n_{cell} with the Fe doping at $T=200$ K for various oxygen contents.

pears to reach a maximum at about $n_{\text{cell}}=1$, while at 100 K the variation is steeper. These results are in good agreement with those of previous studies on Fe-doped 1:2:3.²²

For the $x=0$ series of samples we observed a parabolic-like variation of T_c with n_{cell} at 100 K, whereas at 200 K the variation certainly cannot be described with a single curve. More specifically, even a slight increase in n_{cell} (resulting from the increase of the oxygen content from 6.45 to 6.52) causes T_c to rise from 0 to 40 K, while a further increase in n_{cell} causes a much smaller increase of the T_c . This behavior closely resembles what has been previously reported.³

For the $x=0.05$ series of samples T_c varies linearly with n_{cell} at 100 K, while at 200 K it appears to saturate

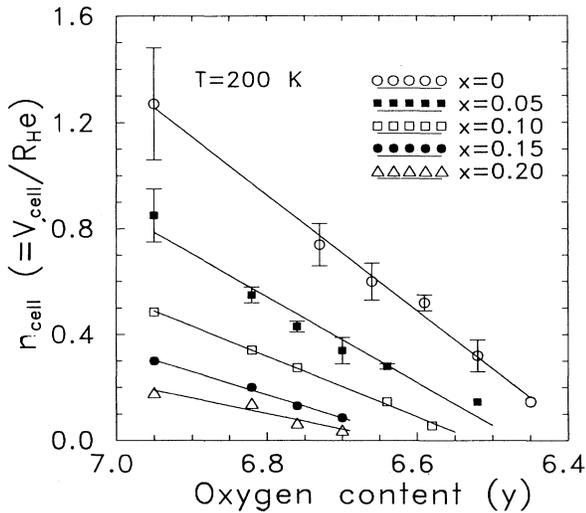


FIG. 5. The variation of n_{cell} with oxygen stoichiometry for all x at $T=200$ K.

at a carrier density near $n_{\text{cell}} \approx 1$. Finally, for the series of samples with $x=0.1, 0.15$, and 0.2 the variation of the T_c with the Hall number is linear at both temperatures. We should also note that the slope dT_c/dn_{cell} increases with increasing iron content, that is, T_c is more sensitive in the Hall number for the Fe-doped rather than for the undoped samples.

The variation of $\cot\theta_H$ with T^2 for the as-prepared samples and for the $x=0.05$ series is shown in Figs. 6(a) and 6(b), respectively. The relation $\cot\theta_H = \alpha T^2 + C$ holds for almost all of our samples. Exceptions are these samples whose resistivity shows a large upturn at low T (an example is the $y=6.64$ sample of the $x=0.05$ series). In these cases the ρ vs T profile determines the $\cot\theta_H$ vs T^2 profile as well.

For the as-prepared Fe-doped samples the slope α , although it does not depend on the Fe content, is doubled with respect to that of the undoped, fully oxygenated 1:2:3. For the $x=0$ sample, $\alpha = 17 \times 10^{-3} \text{ K}^{-2}$,⁹ a value

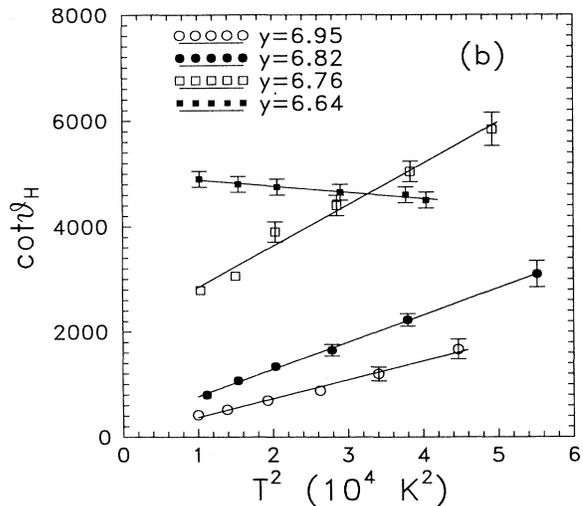
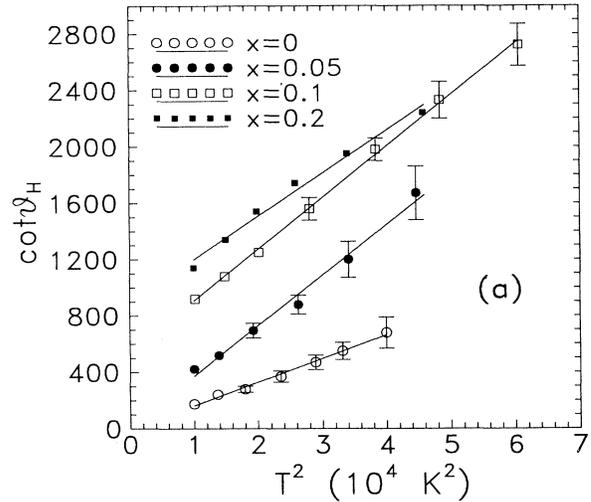


FIG. 6. The variation of $\cot\theta_H$ with T^2 for (a) the as-prepared samples and (b) the $x=0.05$ series.

that is about three times the value of α for single crystals. This difference can be easily explained from the difference in the resistivity values between bulk materials and single crystals.

Regarding the $x = 0.05$ series, the slope α initially increases upon de-oxygenation. As the oxygen content is decreased below 6.70, α begins to decrease gradually since in this region the $\cot\theta_H$ vs T^2 variation is dictated from the ρ vs T variation. In both cases, Fe doping and deoxygenation, C is considerably increased. For the other series of samples the results are completely analogous.

DISCUSSION

Apart from the linear T dependence of n_{cell} which will be discussed in detail below we would like first to comment on the effect that Fe and oxygen doping have on the Hall number.

First, Fe doping causes an exponential decrease in n_{cell} in a similar fashion with Co doping. This is not an unexpected similarity if we recall that both Fe and Co are known to substitute mainly at the Cu(1) chain site.^{24,25} In the case of Co-substituted 1:2:3, this exponential decline in n_{cell} as well as the reduction in the T_c values have been associated with the shortening of the Cu(1)-O(4) bond length as x increases.^{21,26} As O(4) moves away from the Cu(2)-O plane, mobile (plane) holes are transferred to the Cu(1)-O layer where they are localized and therefore do not contribute to the Hall signal and can be measured only chemically.

The idea of dividing the total number of holes into mobile planar holes and localized chain holes was proposed by Tokura *et al.*²⁷ They suggested that there is a threshold at which charge begins to appear on the planes (a reasonable thought since beyond a hole concentration it should be easier to begin to put holes in the planes than to further increase the charge in the chains beyond this value) and also that this threshold charge decreases for smaller oxygen content. Therefore, the role of the chains is simply to act as a charge reservoir while the planar charge is the principal variable governing the superconducting properties. van Woerden and de Leeuw²⁸ explained their data on $\text{Nd}(\text{Ba}_{1-x}\text{Nd}_x)_2\text{Cu}_3\text{O}_{7+\delta}$ on the basis of a redistribution of holes between the planes and the chains, following the model of Tokura *et al.* They have also argued that the driving force for the transfer of holes from the planes to chains is of electrostatic origin. The extra oxygen incorporated at the oxygen chain sites makes the chains more negative and thus more attractive for the positively charged holes.

Miceli *et al.*²⁶ also claimed that there is a critical Cu(1)-O(4) bond length necessary for superconducting behavior but the limited number of data points pose difficulties in accepting such a general point of view. Regarding the $\text{YBa}_2\text{Cu}_{3-x}\text{Fe}_x\text{O}_y$ system, the variation in the Cu(1)-O(4) bond length as x increases is negligible whereas both n_{cell} and T_c vary considerably with x .²⁸ Therefore, it is not clear how the Fe atoms in the Cu(1)-O layer influence the Cu(2)-O plane without significant changes in the distance of the apical oxygen from the Cu(1)-O layer,²⁹ since it is generally accepted that the

Cu(2)-O plane is the structural unit where the essential part of superconductivity is taking place in the high- T_c superconductors.

As for the decrease in n_{cell} caused by deoxygenation, it was found to be linear with y rather than exponential and did not show the plateau behavior observed in the case of the Meissner (and T_c) vs y variation. Regarding the influence of oxygen stoichiometry on the Cu(1)-O(4) bond length in the case of the $\text{YBa}_2\text{Cu}_3\text{O}_y$ system, Jorgensen *et al.*³⁰ reported that it decreases from 1.857 to 1.818 Å as the oxygen content decreases from $y = 6.93$ to $y = 6.45$ and that the variation is linear in the region $6.5 < y < 7$. It has been suggested that the decrease in n_{cell} (as well as in the T_c) upon removing oxygen from the structure is caused by a mechanism analogous to that in the case of Co doping.²¹

Another interesting point is the decrease of the slope dn_{cell}/dT upon doping. For the tetragonal Fe-doped samples ($x \geq 0.1$) and $y < 6.64$, dn_{cell}/dT is practically zero. This zeroing of the slope dn_{cell}/dT is quite important since it can be directly related to the absence of superconductivity in these samples. It is a confirmation of the view that the destruction of the superconducting state and the suppression in the T dependence of the implied carrier density are closely linked. Furthermore, the considerable decrease observed in the slope dn_{cell}/dT upon removing even small amounts of oxygen from Fe-doped samples is further evidence that dn_{cell}/dT and T_c are more closely correlated for the Fe-doped rather than for the undoped 1:2:3.

As for the effect of the Hall number on the T_c it appears that T_c depends strongly on n_{cell} . Regarding the $x = 0$ system, we can distinguish two regions in the T_c vs n_{cell} diagram: one for low oxygen contents ($y < 6.5$) where the increase of the T_c upon increasing y and therefore n_{cell} is very abrupt (in the work of Cooper *et al.*⁴ the T_c increases from 0 to 40 K as y increases from 6.32 to 6.47 while n_{cell} is increased by a factor of 3) and another for $y > 6.5$ where T_c increases gradually with increasing n_{cell} . This comes to terms with the model of Tokura *et al.*²⁷ where the planar charge largely determines T_c and it is the decrease of this charge upon doping that leads to the observed suppression of the T_c . Cava *et al.*³¹ suggested that the decrease of the T_c from 90 to 60 K is due to transfer of (negative) charge of approximately 0.03e/Cu into the planes near $y = 6.6$, and a further charge 0.05e/Cu that is transferred to the planes near $y = 6.45$ results in destroying the superconductivity. Moreover, superconductivity is suppressed more rapidly upon de-oxygenating samples with increasing Fe content, since dT_c/dn_{cell} increases as more iron is incorporated. This result is in excellent agreement with our previous work on Fe-doped 1:2:3,²³ where we have pointed out that as Fe doping increases, the drop in the T_c due to deoxygenation gets more rapid. For both the doped and the undoped system we can therefore conclude that an enhanced Hall number is necessary for the occurrence of a high T_c . Finally, the way in which the carrier density affects T_c depends on the way that the hole density is varied (Fe doping or oxygen depletion). We should no-

tice that Priftis *et al.*²² reported that the variation of the T_c vs n_{cell} (at room temperature) can be fitted with a single curve regardless of the way in which the carrier density is varied.

We turn now to a discussion of the well established $1/T$ dependence of the Hall coefficient R_H which we have also observed in our samples. It is a very unusual result since for a simple metal R_H is temperature independent and is approximately given by the simple relation $R_H = 1/n_H e$. Within conventional Bloch-Boltzmann theory, a temperature-dependent Hall coefficient can arise either when the scattering rate $1/\tau(\mathbf{k})$ is anisotropic with the anisotropy changing with temperature³² or when multiple bands are involved. It was attempted³³ to explain the T dependence observed in 1:2:3 by using a two-band model involving an electron and a hole band. Assuming then that the carrier population is the same in the two bands and that the electron and hole mobilities are dominated by the initial terms of a Taylor expansion in $1/T$ although they differ in second order, the observed $1/T$ dependence of R_H can be easily reproduced. However, the occurrence of such an amazing symmetry between the holes and the electrons for all the cuprates seems at least unrealistic. The independence of R_H on pressure³⁴ as well as the fact that doping increases the value of R_H rather than decreasing it, provide good evidence against multiband models. As for conventional magnetic skew scattering, the absence of saturation at very high magnetic fields and low temperatures [measurements have been performed up to 20 T and down to 2 K in the case of $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ (Ref. 8)] is a good reason for rejecting it as well, as the source of the $1/T$ dependence.

Chien *et al.*³⁵ suggested that in the case of 1:2:3 the only type of carriers are holes and that the observed T dependence of R_H should be closely related to the unusual charge carriers in the Cu(2)-O planes. As for the occurrence of a similar R_H vs T profile in the other superconducting cuprates as well, it might be the case that the anomalous Hall current is probably a universal property of these systems (as are many other properties) and as such it is very unlikely that the details of the Fermi surface and of the phonon spectra can have any influence on the basic electronic state of the Cu(2)-O planes (that are the common structural unit of the cuprates and the place where superconductivity occurs).

Let us now discuss the $\cot\theta_H$ vs T^2 variation. According to Anderson's model, there are two types of quasiparticle excitations, spinons and holons, with different relaxation rates. The transport relaxation rate $1/\tau_{\text{tr}}$ (which determines the resistivity) is related to the spinon-holon scattering that leads to a linear T dependence, i.e., $1/\tau_{\text{tr}} \propto T$. The transverse relaxation rates $1/\tau_H$ is related to the spinon-spinon interaction and therefore varies as T^2 like for any other fermion-fermion interaction. So, $\sigma_{xx} \propto \tau_{\text{tr}} \propto 1/T$ and $\sigma_{xy} \propto \tau_{\text{tr}} \tau_H \propto 1/T^3$. Since $\cot\theta_H (\equiv \sigma_{xx}/\sigma_{xy})$ depends only on τ_H , it varies as T^2 , that is, $\cot\theta_H = \alpha T^2 + C$. The slope α is related to the reciprocal spinon bandwidth and C is a measure of the in-plane magnetic impurity scattering rate.

In the case of c -axis-oriented thin films with various oxygen contents,¹⁷ the observed increase in the slope α upon de-oxygenation has been related to the reduction of the true carrier density (which also results in decreasing the spinon bandwidth as well). For the sintered samples we studied, the tensor nature of the Hall coefficient precludes a direct comparison with the slope α . Nevertheless, it is worth noticing that the increase we observed in the slope α upon Fe doping is consistent with such a picture, since Fe decreases the carrier density. However, recent experiments in Co-doped single crystals¹¹ have shown an opposite trend: α decreases upon Co doping.

The increase of α observed in the $x=0.05$ series upon removing oxygen from the system (we are referring to the $y \geq 6.70$ samples) can also be interpreted in the same context. Here, we should also mention that the variation of α upon deoxygenation of the undoped 1:2:3 is not well established. Experiments in c -axis-oriented thin films have shown two different behaviors: α increases as y decreases¹⁷ and α decreases as y decreases and saturates in the 60 K plateau region.¹⁸ Probably experiments in single crystals are needed to resolve this uncertainty.

The increase of C both upon Fe doping and de-oxygenation can, to a certain extent, be attributed to disorder scattering but it is more likely that the reason is the largely increased resistivity values. It certainly cannot be explained with the creation of local magnetic moments in the CuO_2 planes, although a small fraction of the Fe atoms can occupy the Cu(2) sites.

CONCLUSIONS

We have studied extensively the variation of the Hall number with Fe doping ($0 \leq x \leq 0.2$), oxygen content ($6 < y < 7$), as well as with temperature (100–250 K) for the $\text{YBa}_2\text{Cu}_{3-x}\text{Fe}_x\text{O}_y$ system. Both Fe doping and oxygen depletion decrease n_{cell} but in a different way (at least up to 200 K.) The exponential decrease in n_{cell} with increasing x becomes more rapid as the oxygen content decreases. The slope dn_{cell}/dT and the T_c are closely related since the zeroing of the slope coincides with the absence of superconductivity for $y < 6.64$ and $x \geq 0.1$. T_c is a sensitive function of the Hall number for both the undoped and the Fe-doped system. Especially for the Fe-doped samples, dT_c/dn_{cell} (the hole density is varied through de-oxygenation) increases with increasing Fe content. The $\cot\theta_H$ vs T^2 relation holds for the majority of our samples. The variation of the parameters α and C with Fe doping and oxygen content is discussed, within the reservations posed by the polycrystallinity of our samples, and it is compared with experiments in c -axis-oriented thin films and single crystals.

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- ¹S. W. Tozer, A. W. Kleinsasser, T. Penney, D. Kaiser, and F. Holtzberg, *Phys. Rev. Lett.* **59**, 1768 (1987).
- ²S. W. Cheong, S. E. Brown, Z. Fisk, R. S. Kwok, J. D. Thompson, E. Zirngieble, G. Gruner, D. E. Peterson, G. L. Wells, R. B. Schwarz, and J. R. Cooper, *Phys. Rev. B* **36**, 3913 (1987).
- ³Z. Z. Wang, J. Clayhold, N. P. Ong, J. M. Tarascon, L. H. Greene, W. R. McKinnon, and G. W. Hull, *Phys. Rev. B* **36**, 7222 (1987).
- ⁴J. R. Cooper, S. D. Obertelli, A. Carrington, and J. W. Loram, *Phys. Rev. B* **44**, 12 086 (1991).
- ⁵For a review see N. P. Ong, in *Physical Properties of High-Temperature Superconductors*, edited by D. M. Ginsberg (World Scientific, Singapore, 1990), Vol. 2, p. 459.
- ⁶M. Suzuki, *Phys. Rev. B* **39**, 2312 (1989).
- ⁷Y. Kubo, Y. Shimikawa, T. Manako, and H. Igahashi (unpublished).
- ⁸Z. Z. Wang, T. R. Chien, N. P. Ong, J. M. Tarascon, and E. Wang, *Phys. Rev. B* **43**, 3020 (1991).
- ⁹T. R. Chien, Z. Z. Wang, and N. P. Ong, *Phys. Rev. Lett.* **67**, 2088 (1991).
- ¹⁰P. W. Anderson, *Phys. Rev. Lett.* **67**, 2092 (1991).
- ¹¹A. Carrington, A. P. Mackenzie, C. T. Lin, and J. R. Cooper, *Phys. Rev. Lett.* **69**, 2855 (1991).
- ¹²W. E. Pickett, *Rev. Mod. Phys.* **61**, 433 (1989); W. E. Pickett, H. Krakauer, R. E. Cohen, and D. J. Singh, *Science* **255**, 46 (1992).
- ¹³Y. Kubo and T. Manako, *Physica C* **197**, 378 (1992).
- ¹⁴P. S. Wang, J. C. Williams, K. D. D. Rathnayaka, B. D. Hennings, D. G. Naugle, and A. B. Kaiser, *Phys. Rev. B* **47**, 1119 (1993).
- ¹⁵N. P. Ong, T. W. Jing, T. R. Chien, Z. Z. Wang, T. V. Ramakrishnan, J. M. Tarascon, and K. Remschnig, *Physica C* **185-189**, 34 (1991).
- ¹⁶Wu Jiang, L. J. Peng, S. J. Hagen, and R. L. Greene, *Phys. Rev. B* **46**, 8694 (1992).
- ¹⁷P. Xiong, G. Xiao, and X. D. Wu, *Phys. Rev. B* **47**, 5516 (1993).
- ¹⁸B. Wuyts, E. Osquiguil, M. Maenhoudt, S. Libbrecht, Z. X. Gao, and Y. Bruynseraede, *Phys. Rev. B* **47**, 5512 (1993).
- ¹⁹G. Xiao, P. Xiong, and M. Z. Cieplak, *Phys. Rev. B* **46**, 8687 (1992).
- ²⁰He Zhenhui, Chen Zuyao, Zhao Zhanchuen, Xu Zhuan, Qian Yitai, and Zhang Qirui, *Phys. Lett. A* **132**, 217 (1988).
- ²¹J. Clayhold, S. Hagen, Z. Z. Wang, N. P. Ong, J. M. Tarascon, and P. Bardoux, *Phys. Rev. B* **39**, 777 (1989); **39**, 7324 (1989).
- ²²G. Priftis, A. M. Ghopayeb, O. Gorochoy, R. Suryanarayanan, H. Pankowska, and M. Ratdau, *Physica C* **162-164**, 1201 (1989).
- ²³G. Kallias, V. Psyharis, D. Niarchos, and M. Pissas, *Physica C* **174**, 316 (1991).
- ²⁴J. M. Tarascon, P. Bardoux, P. F. Miceli, L. H. Greene, G. W. Hull, M. Eibschutz, and S. A. Sunshine, *Phys. Rev. B* **37**, 7458 (1988).
- ²⁵R. Sonntag, D. Hohlwein, A. Hoser, W. Prandl, W. Schafer, R. Kiemel, S. Kemmler-Sack, S. Losch, M. Schlichenmaier, and A. W. Hewat, *Physica C* **159**, 141 (1989).
- ²⁶P. F. Miceli, J. M. Tarascon, L. H. Greene, P. Bardoux, F. J. Rotella, and J. D. Jorgensen, *Phys. Rev. B* **37**, 5932 (1988).
- ²⁷Y. Tokura, J. B. Torrance, T. C. Huang, and A. I. Nazzari, *Phys. Rev. B* **38**, 7156 (1988).
- ²⁸R. A. M. van Woerden and D. M. de Leeuw, *Physica C* **165**, 221 (1990).
- ²⁹Y. Xu, M. Suenaga, J. Taftø, R. L. Sabatini, A. R. Moodenbaugh, and P. Zolliker, *Phys. Rev. B* **39**, 6667 (1989).
- ³⁰J. D. Jorgensen, B. W. Veal, A. P. Paulikas, L. J. Nowicki, G. W. Crabtree, H. Claus, and K. W. Kwok, *Phys. Rev. B* **41**, 1863 (1990).
- ³¹R. J. Cava, A. W. Hewat, E. A. Hewat, B. Batlogg, M. Marezio, K. M. Rabe, J. J. Krajewski, W. F. Peck, Jr., and L. W. Rupp, Jr., *Physica C* **165**, 419 (1990).
- ³²J. Ziman, *Phys. Rev. B* **121**, 1320 (1961).
- ³³A. Davidson, P. Santhanam, A. Palevski, and M. J. Brady, *Phys. Rev. B* **38**, 3828 (1988).
- ³⁴I. D. Parker and R. H. Friend, *J. Phys. C* **21**, L345 (1988).
- ³⁵T. R. Chien, D. A. Brawner, Z. Z. Wang, and N. P. Ong, *Phys. Rev. B* **43**, 6242 (1991).