RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 46, NUMBER 13 1 OCTOBER 1992-I

Universal Hall effect in $La_{1.85}Sr_{0.15}Cu_{1-x}A_xO_4$ systems (A = Fe,Co,Ni,Zn,Ga)

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(Received 8 June 1992)

The temperature dependence of the Hall angle is shown to follow $\cot\Theta_H = \alpha T^2 + C(x)$ in five differently doped $La_{1.85}Sr_{0.15}Cu_{1-x}A_xO_4$ systems (x is the impurity concentration and A = Fe, Co, Ni, Zn, or Ga). The slope α remains approximately constant with doping, regardless of the different magnetic or valence state of an impurity. The quantity $C(x)$ increases linearly with x. These observations are consistent with the Hall effect in the $YBa_2(Cu_{1-x}Zn_x)_3O_7$ system, and in conformity with Anderson's theory of the Hall effect in a Luttinger liquid.

One of the important signatures of the normal state of cuprate superconductors is the temperature dependence of the Hall coefficient (R_H).[1] The observed T dependence is inconsistent with the Fermi-liquid description of a single energy band. Because of its close relevance to the normal state, extensive experimental and theoretical studies have been carried out to clarify the origin of the Hall effect.[1] Some attempts[2–4] have involved a Fermi-liquid description with modifications such as the introduction of multibands, magnetic skew scattering, etc. These attempts have required many fitting parameters or have depended on a delicate balance between competing effects. As a result the universal nature of the Hall effect in the cuprates has tended to be overlooked. A nonconventional approach to tackle the anomalous Hall effect has started to gain more attention.[5–7] Recently, an analysis[8] revealed that the cotangent of the Hall angle ($\cot\Theta_H$) best characterizes the anomalous Hall effect in $YBa_2(Cu_{1-x}Zn_x)_3O_7$. It was found that

$$\cot\Theta_H = \alpha T^2 + C(x) \quad (1)$$

from 95 K to about 240 K. What is most striking is that the slope α is constant as x (Zn doping on Cu sites) varies, even though the T dependence of R_H changes substantially. The only effect of the Zn doping is to vary the parameter C proportionally to x. Anderson[7] has proposed an explanation for this finding in the framework of the Luttinger-liquid theory. The behavior of $\cot\Theta_H$ is attributed to the intrinsic relaxation rate of the elementary excitations (the spinons) in the normal state. Among the appealing features of the theory are that few parameters are required to describe the normal-state transport, and that the understanding of the general features of the normal-state properties does not involve the details of the electronic structure.

Confirmation of a universal $\cot\Theta$ vs T^2 dependence in other cuprate superconductors would provide strong support for the theory. The $La_{1.85}Sr_{0.15}Cu_{1-x}A_xO_4$ (2:1:4) system, where A represents various magnetic or nonmagnetic impurities, is an excellent candidate for such a study. Contrary to the case of the $YBa_2Cu_3O_7$ (1:2:3) system in which the presence of Cu-O chains complicates the interpretation of the doping studies, the electrical transport in the 2:1:4 system is dominated by the CuO_2 planes. The magnetic state of various impurities and their effect on superconductivity have been well characterized in the 2:1:4 system.[9,10] Here we present a systematic study of the Hall effect in five 2:1:4 series with Zn, Ga, Fe, Co, and Ni impurities. The $\cot\Theta_H$ vs T^2 relation is confirmed in each series with an intercept C proportional to x and a value of the slope α which is close to being *constant*, except for the case of Zn where the slope increases slightly.

The samples used for this study were made under identical conditions using a standard solid-state reaction method. The impurity doping level is limited to $x \leq 0.06$ and all samples are single phase as confirmed by x-ray diffraction. In fact a doping level as high as 0.3 (for Zn and Ni) or 0.1 (for Fe) can be achieved without the formation of any secondary phases. We have used these samples previously to study the local magnetic moment of the impurities[9] and their effect on superconductivity and the normal state.[10,11] To measure the Hall effect a five-probe arrangement was used on regularly shaped samples ($\sim 15 \times 3 \times 0.3$ mm^3). The Hall voltage was obtained with a measuring current of 100 mA and by sweeping the magnetic field from +8 T to −8 T. For most of the samples measurements were carried out in the temperature range of 80–300 K. We have also checked the magnetic-field dependence of R_H for every series. In the temperature range of our study R_H is linear up to our maximum field of 8 T, with no indication of the saturation in R_H that would be expected from magnetic skew scattering.[12] We remind that our samples are in ceramic form. It would be preferable to have single crystals. However, it is very difficult to synthesize homogeneous single crystals of the 2:1:4 series with both high-level Sr doping and impurity doping on the Cu site. Fortunately, the temperature dependence of resistivity and Hall effect are the same for both single crystals and ceramic samples, with difference only in the magnitude of resistivity. We have taken this factor into consideration when analyzing our data.

Measurements of the T dependence of the resistivity for the five series are published elsewhere.[10,11] As the impurity content is increased the linear $\rho(T)$ in pure parent material gives way to the upturn at low T. The critical doping level, x_c, at which T_c becomes zero, ranges from 1.8 at.% for Fe, to about 2.6 at.% for Zn, Ga, and Co, and 4.4 at.% for Ni. The values of x_c are correlated with the size of the magnetic moment of each impurity,[9] a clear indication that a magnetic pair-breaking effect is the dominant mechanism in suppressing superconductivity.

The T dependence of $1/R_H$ for the five series is shown in Fig. 1. There is a substantial T dependence in every sample, but it becomes smaller as x increases. A similar effect has been observed in 1:2:3 systems doped with Zn or Co.[13] The case of Zn doping is exceptional in that Zn doping increases the Hall number $n_H = 1/(eR_H)$, while the opposite is seen in all other cases. Both Zn and Ni are divalent (like Cu) and their doping in the 2:1:4 system does not change the oxygen content. However, $1/R_H$ is affected differently in these two systems. Another interesting observation is that even dilute doping causes a substantial variation in $1/R_H$ in all series. For example, doping with 3 at.% Co changes $1/R_H$ by as much as 40%.

To extract the underlying controlling parameters for the Hall-effect results we adopted the Hall angle analysis which has successfully unified the anomalous T dependence in the $YBa_2(Cu_{1-x}Zn_x)_3O_7$ system.[8] In Fig. 2 we show $\cot\Theta_H$ as a function of T^2 for the five 2:1:4 series. $\cot\Theta_H$ is the ratio of the longitudinal voltage to the transverse (Hall) voltage, i.e., $\cot\Theta_H = \rho_{ab}/R_H B$, where ρ_{ab} is the average resistivity in the a-b plane. It may be seen that Eq. (1) is well satisfied in the temperature range from about 100 to 300 K. More importantly, the slope α is close to *constant* for every series except the Zn series. Another exception is the sample with 2.7% Fe doping, whose slope differs from the rest. This may be because it is a nonideal sample, or because the sample is close to the metal-insulator transition. It is remarkable that while the T dependence of $\rho(T)$ and $R_H(T)$ changes substantially as the doping level is increased, $\cot\Theta_H$, the ratio of $\rho(T)$ to $R_H(T)$, strictly follows the simple T^2 dependence. In the case of Zn the slope α increases about 20% with doping, in contrast with the constant slope observed in the Zn-doped 1:2:3 system. The cause of the increase is not clear and further study is underway.

In the conventional Drude model, $\cot\Theta_H = (\omega\tau)^{-1}$, where ω is the cyclotron frequency and τ the quasiparticle lifetime. At high temperatures where the electron-phonon interaction dominates, one would expect a linear T dependence of $\cot\Theta_H$. This is not the case in any of our systems. Anderson[7] argues that in cuprate superconductors the relaxation rate τ_H^{-1} (measured by $\cot\Theta_H$) is due to the spinon-spinon interaction and the spinon–magnetic-impurity scattering (Matthessen's rule). Scattering between spinons is a T^2 process as in any other fermion-fermion interaction. The spinon–magnetic-impurity scattering is a T-independent process, only adding a constant to $\cot\Theta_H$. According to Anderson the slope α in Eq. (1) should be proportional to $1/J$ (where J is the exchange interaction) or to the reciprocal of the spinon bandwidth. In all of our systems α remains approximately constant, indicating that the exchange interaction J does not change for the different impurities. We have found earlier[9] that Fe carries the largest mo-

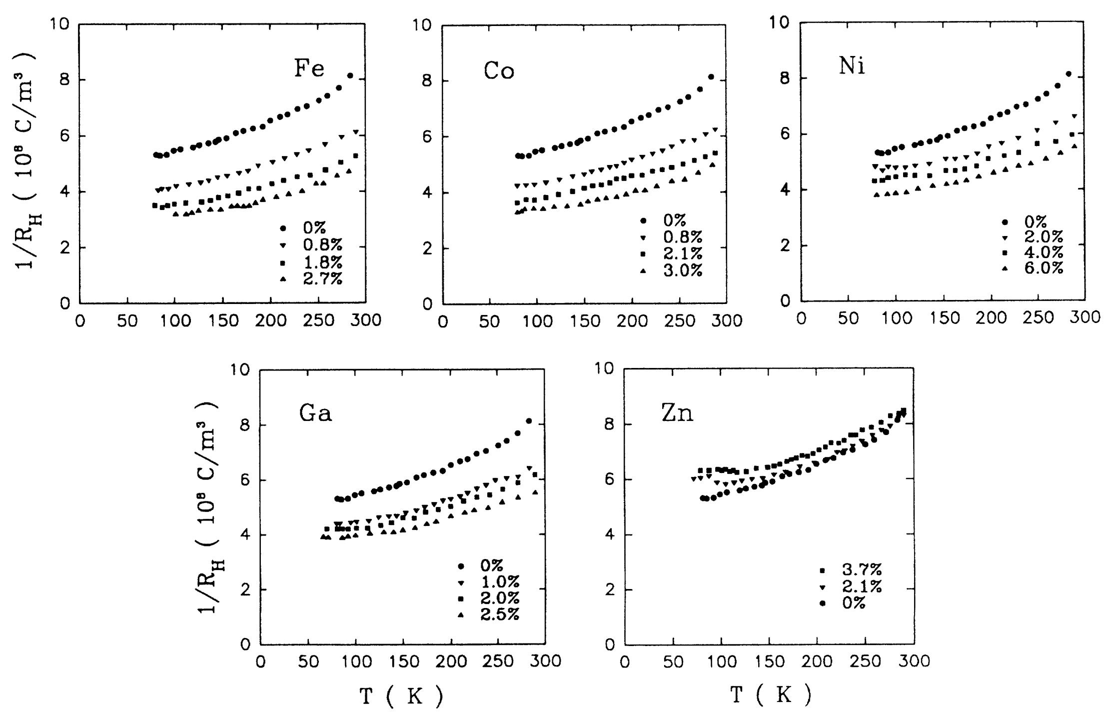

FIG. 1. Temperature dependence of $1/R_H$ in five $La_{1.85}Sr_{0.15}Cu_{1-x}A_xO_4$ systems (A = Fe, Co, Ni, Zn, and Ga).

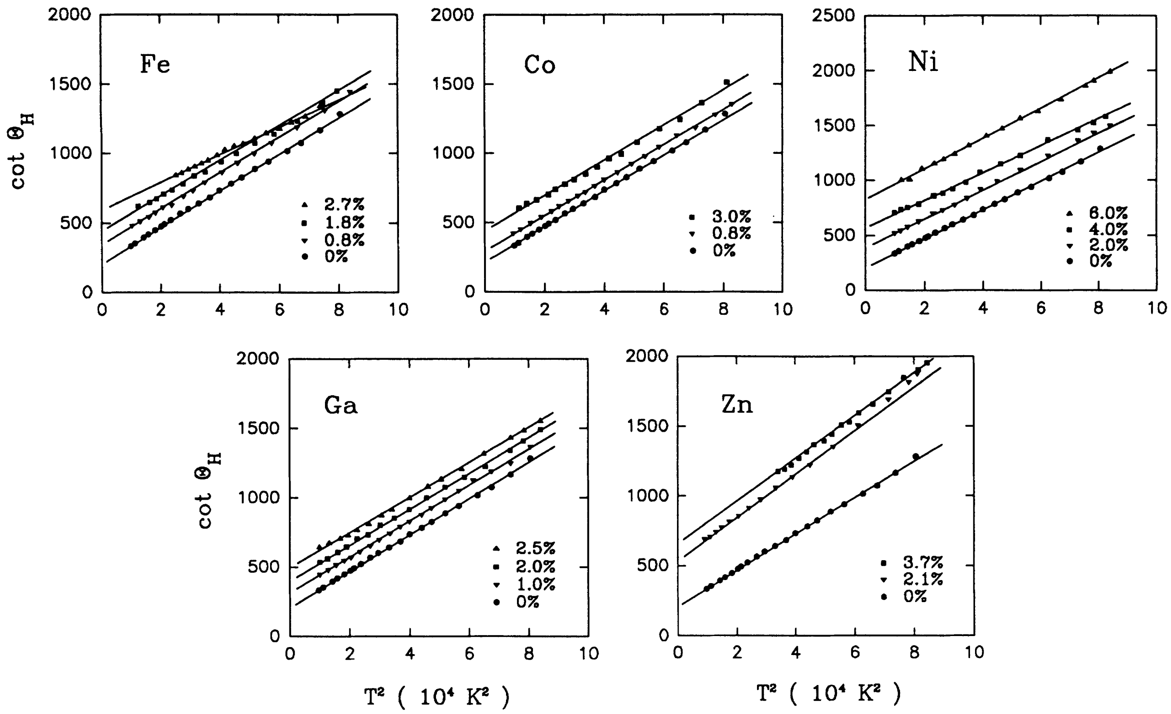

FIG. 2. $\cot\Theta_H$ as a function of T^2 for $La_{1.85}Sr_{0.15}Cu_{1-x}A_xO_4$ in 8 T.

ment of $5\mu_B$ and Ni carries the lowest moment of $0.6\mu_B$. Zn, Ga, and Co, though nonmagnetic by themselves (Co is in a zero-spin state), induce a magnetic moment of about $1\mu_B$ in the Cu-O_2 plane. Clearly the value of the impurity moment has little effect on J.

The dominant effect of the impurities is to change the constant $C(x)$ in Eq. (1). In Anderson's theory $C(x)$ is proportional to the impurity concentration x. As shown in Fig. 3, C indeed increases linearly with x. Fe has the largest magnetic moment and tends to increase C more than the other impurities. It is interesting to note that C is appreciable even without impurities, i.e., in undoped $La_{1.85}Sr_{0.15}CuO_4$. For undoped $YBa_2Cu_3O_7$, C is close to zero. This may be because of the intrinsic disorder due to Sr doping, which affects the neighboring Cu-O_2 local structure.

The average value of α for our polycrystalline samples is $1.33\times10^{-2}\,K^{-2}$ at 8 T. It is known that a single crystal and a polycrystalline sample share basically the same T dependence of $1/R_H$ and $\rho(T)$.[1] The values of $1/R_H$ are within 10% of each other, and the value of ρ is about 5.5 times larger in polycrystals[10,11] than in single crystals

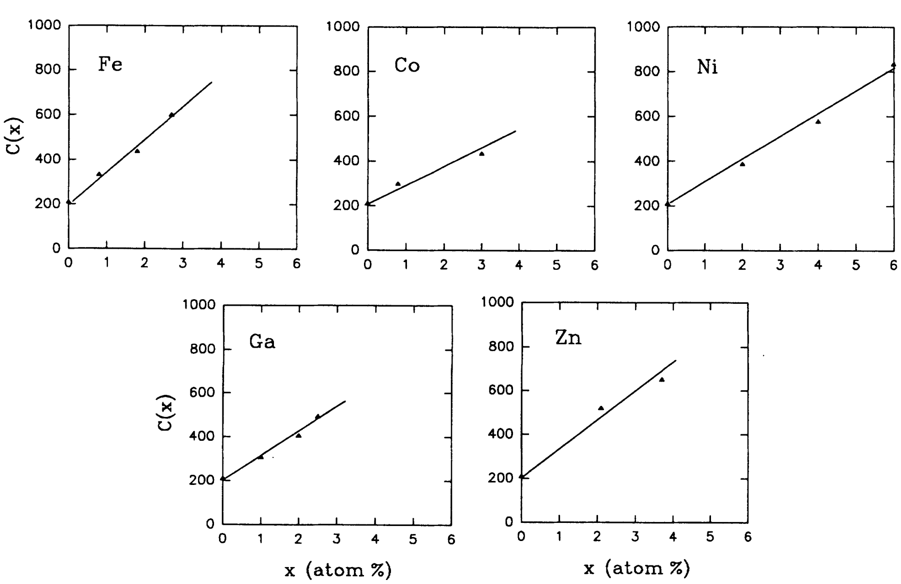

FIG. 3. $C(x)$ in $\cot\Theta_H = \alpha T^2 + C(x)$ as a function of impurity concentration x in $La_{1.85}Sr_{0.15}Cu_{1-x}A_xO_4$ in 8 T.

or epitaxial films.[14] α should therefore be scaled down to about $2.4 \times 10^{-3}\,\mathrm{K}^{-2}$ for a 2:1:4 single crystal. In a 1:2:3 single crystal $\alpha = 5.11 \times 10^{-3}\,\mathrm{K}^{-2}$,[8] so that α has the same order of magnitude for both systems. This is reasonable because J is comparable in both the 1:2:3 and 2:1:4 systems. The parameter C for undoped 2:1:4 is about 210. It may be expected to be about 40 for a single crystal, which is substantially higher than for undoped 1:2:3 where C is about 5. The average slope dC/dx is about 10 000 for our polycrystalline samples, and reduces to about 1900 for a 2:1:4 single crystal (compared to 2300 in a 1:2:3 single crystal).

The universal nature of the Hall effect in all five systems is the highlight of this work. As mentioned earlier the magnetic states of the various impurities differ a great deal. Because of their different valence states they have different effects on the carrier concentration. Nevertheless the T dependence of $\cot\Theta_H$ is remarkably invariant with impurity doping. This results naturally from Anderson's theory, because the relaxation rate $1/\tau_{tr}$ of the holon excitations cancels out in $\cot\Theta_H$. The only relevant relaxation rate is that of the spinon excitations $(1/\tau_H)$, which is closely related to the presence of a magnetic impurity. And on Cu-O_2 planes, every impurity (Fe, Co, Ni, Zn, or Ga) is *magnetic* in nature,[9] a property highly characteristic of the Cu-O_2 planes.

We thank A. Houghton, B. Marston, and P. Lindenfeld for helpful discussions. This work was supported by NSF Grant Nos. DMR 9024402 and DMR 9022033 and by a grant from the Polish Committee for Scientific Research. One of the authors (G.X.) wishes to acknowledge partial financial support from the A. P. Sloan Foundation.

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