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Hall effect in Zn-doped $YBa_2Cu_3O_{7-\delta}$ revisited: Hall angle and the pseudogap

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The temperature dependence of the Hall coefficient is measured with high accuracy in a series of $YBa_2(Cu_{1-z}Zn_z)_3O_{6.78}$ crystals with $0 \leqslant z \leqslant 0.013$. We found that the cotangent of the Hall angle, $\cot \theta_H$, starts to deviate upward from the T^2 dependence below T_0 (~ 130 K), regardless of the Zn concentration. We discuss that this deviation is caused by the pseudogap; the direction of the deviation and its insensitivity to the Zn doping suggest that the pseudogap affects $\cot \theta_H$ through a change in the effective mass, rather than through a change in the Hall scattering rate. [S0163-1829(99)50346-9]

The strong temperature dependence of the Hall coefficient R_H of the high-T_c cuprates has been considered to be one of the most peculiar properties of their unusual normal state.[1] The rather complex behavior[2] of $R_H(T)$ can be turned into a simpler one by looking at the cotangent of the Hall angle,[3] $\cot \theta_H \equiv \rho_{xx}/\rho_{xy}$; it has been shown that $\cot \theta_H$ of cuprates behaves approximately as T^2, regardless of material[4] and carrier concentration.[2] This remarkable simplicity in the behavior of $\cot \theta_H$ led to the idea[3,5] that $\cot \theta_H$ reflects a Hall scattering rate τ_H^{-1}, which is different from the scattering rate τ_{tr}^{-1} governing the diagonal resistivity ρ_{xx}. There are two physical pictures to account for this apparent separation of the scattering rates: One picture considers that two distinct scattering times τ_{tr} and τ_H, possibly associated with different particles, govern different kinds of scattering events.[5,6] The other picture considers that the scattering time is strongly dependent on the position on the Fermi surface (FS) and that ρ_{xx} and $\cot \theta_H$ are governed by the scattering events on different parts of the FS.[2,7]

Separate from the above development, it has become a common understanding[8,9] that in underdoped cuprates a pseudogap in the density of low-energy excitations is developed at a temperature much higher than the superconducting transition temperature T_c. In underdoped $YBa_2Cu_3O_{7-\delta}$ (YBCO), the in-plane resistivity ρ_{ab} shows a clear downward deviation from the T-linear behavior below a temperature T^*, which has been discussed to mark the onset of the pseudogap.[10] This T^* is notably higher than the other characteristic temperature T_g determined from the onset of a suppression in the Cu NMR relaxation rate,[11] which has also been associated with the pseudogap. The presence of two different temperature scales, T^* and T_g, is intriguing. It was proposed recently that at the upper temperature scale T^* the CuO_2 plane starts to develop local antiferromagnetic correlations[8] or charged stripe correlations;[12] the lower temperature scale T_g corresponds to the opening of a more robust pseudogap in the density of states,[8] which can be observed by the angle-resolved photoemission[13] or by the tunneling spectroscopy.[14]

It was previously discussed[10] that the pseudogap causes a deviation from the T^{-1} behavior in $R_H(T)$ at T^*. The conspiring changes in $\rho_{ab}(T)$ and $R_H(T)$ at T^* leave the T^2 behavior of $\cot \theta_H$ unchanged at T^*, which led to the belief that $\cot \theta_H$ is rather insensitive to the opening of the pseudogap. However, given the recent understanding that the pseudogap has two characteristic temperatures T^* and T_g, it is left to be investigated how $\cot \theta_H(T)$ behaves around T_g.

Since the pseudogap effect is expected to be related to the antiferromagnetic fluctuations,[8] there have been efforts to investigate how the pseudogap feature is affected by Zn doping onto the CuO_2 planes, which produces spin vacancies. The reported Zn-doping effects on the pseudogap are not simple; for example, the pseudogap feature in $\rho_{ab}(T)$ in underdoped YBCO crystals is almost unchanged,[15] while the suppression in the Cu NMR relaxation rate below T_g is diminished with only 1% of Zn.[16] To build a complete picture of the pseudogap effect, it is also useful to investigate how the Zn doping affects the pseudogap in the Hall channel.

In this paper, we report the results of our measurements of the Hall effect in $YBa_2(Cu_{1-z}Zn_z)_3O_y$ crystals with $y = 6.78$, which corresponds to an underdoped concentration. At this composition $y = 6.78$, which gives $T_c \simeq 75$ K in pure crystals, a peak in $R_H(T)$ can be clearly seen and also the pseudogap feature in $\rho_{ab}(T)$ is clearly discernible (due to the rather wide T-linear region above T^*); from the literature, we can infer that T^* is about 200 K (Ref. 10) and T_g is about 130 K (Ref. 17). Our measurements of three samples with different Zn concentrations ($z = 0$, 0.006, and 0.013) found that a deviation from the T^2 behavior in $\cot \theta_H$ takes place in all the samples at the same temperature T_0 which is very close to T_g, indicating that the pseudogap indeed affects cot θ_H near T_g and that the effect is robust against Zn doping.

There have been several publications reporting the effect of Zn doping on R_H in YBCO, but the results are not converged. The data by Chien, Wang, and Ong indicate that R_H of optimally doped crystals increases with increasing z in the whole temperature range above T_c and the T dependence becomes less pronounced[3] [it is possible that in their samples the effective carrier concentration is changing, because the slope of $\rho_{ab}(T)$ is increasing with z]. Mizuhashi *et al.* reported that R_H increases over the whole temperature range with z (almost like a parallel shifting), while the slope of $\rho_{ab}(T)$ in the T-linear part is unchanged.[15] On the other hand, Walker, Mackenzie, and Cooper reported that, in their Zn-doped crystalline thin films, R_H at 300 K remains essentially unchanged, while at low temperatures R_H is progressively suppressed with increasing z.[2,18] In the present work, we therefore paid particular attention to reduce the errors in the measurement of R_H; the Hall voltage is measured with magnetic-field sweeps at constant temperatures, and errors due to the geometrical factors are minimized by making small voltage contacts and by determining the sample thickness with a high accuracy. We note that making the voltage contacts on the side faces (not on the top face) of the crystals is essential in reducing the error and increasing the reproducibility.

The Zn-doped YBCO single crystals are grown by a flux method using pure Y_2O_3 crucibles.[19] All the crystals measured here are naturally twinned. The oxygen content is tuned to $y=6.78$ by annealing the crystals with pure YBCO powders in air at 575 °C for 37 h, and subsequent quenching to room temperature. The final oxygen content is confirmed by iodometric titration. The actual Zn concentration in the crystals is measured with the inductively coupled plasma (ICP) spectrometry with an error in z of less than ± 0.001.

The measurements are performed with a low-frequency (16 Hz) ac technique. Longitudinal and transverse voltages are measured simultaneously using two lock-in amplifiers during the field sweeps at constant temperatures. For the transverse signal, we achieved a high sensitivity by subtracting the offset voltage at zero field (the offset comes from a slight longitudinal misalignment between the two Hall voltage contacts). The temperature is stabilized using a high-resolution resistance bridge with a Cernox resistance thermometer. We confined the maximum magnetic field to 4 T, within which the error of the Cernox thermometer caused by its own magnetoresistance is negligibly small in the temperature range of the present study. The magnetic field is applied along the c axis of the crystals. To enhance the temperature stability, the sample and the thermometer are placed in a vacuum can with a weak thermal link to the outside. The achieved stability in temperature during the field sweeps is better than a few mK. The data are taken from -4 T to $+4$ T, and then the asymmetrical component is calculated to obtain the true Hall voltage. The final accuracy in the magnitude of R_H and ρ_{ab} reported here is estimated to be better than $\pm 5\%$, and the relative error in the data for each sample is less than $\pm 2\%$.

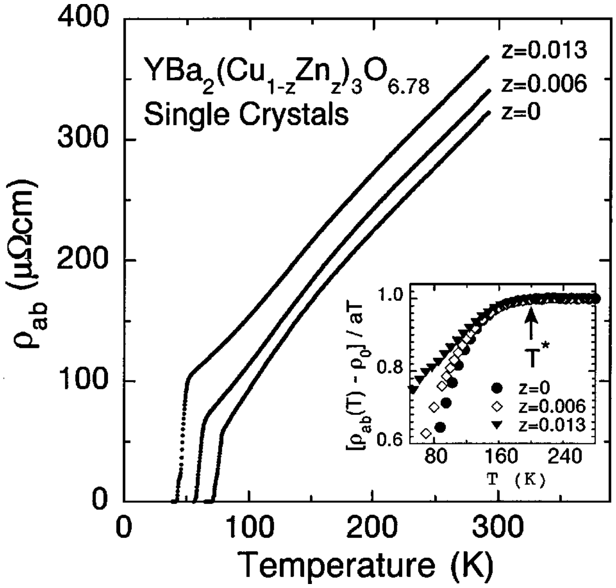

FIG. 1. T dependence of ρ_{ab} for the pure and Zn-doped samples. Inset: Plots of $[\rho_{ab}(T)-\rho_0]/aT$ vs T, where $\rho_0=13.9$, 34.6, and 63.4 $\mu\Omega$ cm for $z=0$, 0.006, and 0.013, respectively. The slope $a(=1.05)$ is unchanged with z. T^* is marked by an arrow.

Figure 1 shows the temperature dependence of ρ_{ab} for the three Zn concentrations. Above ~200 K, ρ_{ab} of all the three samples shows a good T-linear behavior and the slope of this T-linear part does not change with z. As shown in the inset to Fig. 1, a downward deviation from the T-linear dependence takes place at the same temperature for all three samples, indicating that the upper pseudogap temperature T^* does not change with z. This result is in good agreement with the previous reports.[15,18]

Figure 2 shows the temperature dependence of R_H for the three samples. Our results are somewhat different from previous results on single crystals,[3,15] but rather resemble that of the thin film result.[18] Notably, R_H around 250 K does not change with z, while the peak at 110 K is clearly suppressed with increasing Zn concentration. Still, the behavior of cot θ_H is in good agreement with the previous studies; as is shown

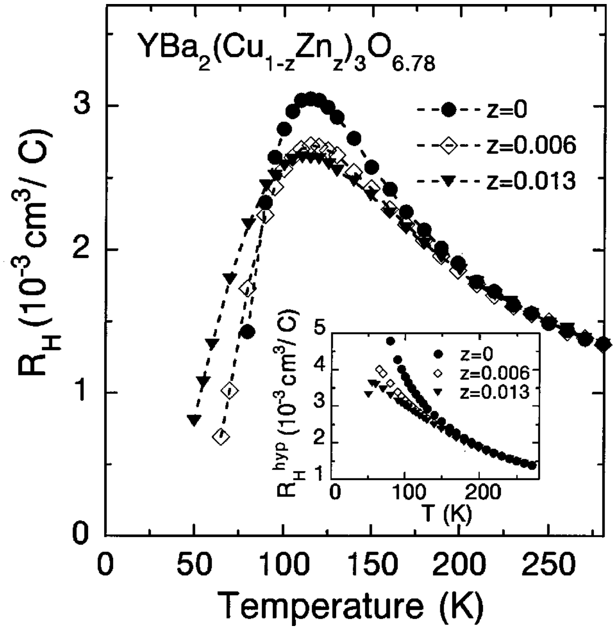

FIG. 2. T dependence of R_H for pure and Zn-doped samples. Inset: Plot of R_H^{hyp} vs T for the three samples, see text.

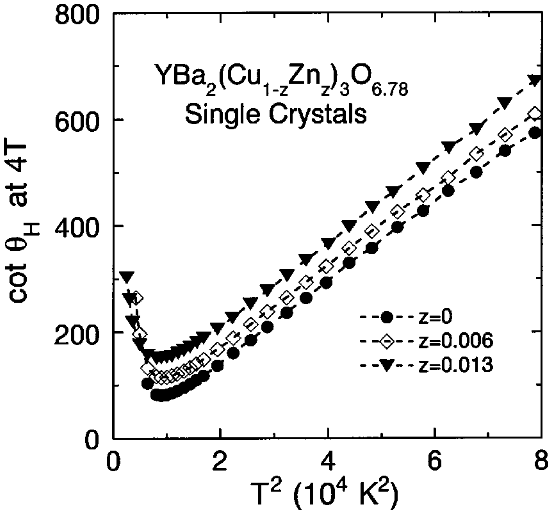

FIG. 3. Plots of cot θ_H vs T^2 for the three samples.

in Fig. 3, cot θ_H changes approximately as T^2 in a rather wide range, and the Zn impurities add a T-independent offset which is roughly proportional to z.

We note that the Zn-doping effect on $R_H(T)$ observed here is naturally expected in the context of the two scattering time scenario. One can infer that the primary effect of Zn-doping is to add some constant impurity-scattering rates to both τ_{tr}^{-1} and τ_H^{-1}, because both $\rho_{ab}(T)$ and cot $\theta_H(T)$ show essentially parallel shifts upon Zn doping. Since one can approximately express $\tau_{tr}^{-1} \sim T$ and $\tau_H^{-1} \sim T^2$ in pure samples, the scattering rates in Zn-doped samples can be approximated as $\tau_{tr}^{-1} \sim T+A$ and $\tau_H^{-1} \sim T^2+B$. From the relation $R_H H = \rho_{ab}/\cot\theta_H \sim \tau_H/\tau_{tr}$, R_H is approximately written as $R_H \sim (T+A)/(T^2+B)$ in Zn-doped samples. If we compare this expression with that for the pure samples, $R_H^{pure} \sim T/T^2 \sim T^{-1}$, we can infer that at high temperatures R_H in Zn-doped sample should approach R_H^{pure}, while at low temperatures R_H in Zn-doped sample is expected to become smaller than R_H^{pure} (which can be easily seen when one considers $T \rightarrow 0$). The above heuristic argument implies that the weakening of the T dependence of $R_H(T)$, combined with a z-independent room-temperature R_H, is a rather natural consequence of the Zn doping in the two scattering time scenario, although this effect has not been well documented before.

Now let us analyze the data in more detail in regard to the T dependence of cot θ_H. A close examination of Fig. 3 tells us that the data for $z=0$ and 0.006 are slightly curved in this plot; we found that the best power laws to describe the data in a wide temperature range are $T^{1.85}$, $T^{1.9}$, and $T^{2.0}$, for $z=0$, 0.006, and 0.013, respectively. In Fig. 4, we show plots of $(\cot\theta_H - C)/T^{\alpha}$ vs T, which cancels out the power-law temperature dependence and therefore we can easily see the temperature range for the T^{α} dependence to hold well. Here, C is the offset value (which increases with z) and α is the best power for each Zn concentration. It is clear from Fig. 4 that the power-law temperature dependence of cot θ_H holds very well down to a temperature T_0 ($\sim$130 K) and then starts to deviate in all the three samples. Incidentally, the deviation occurs at a temperature very close to T_g, which is $\sim$130 K for $y \approx 6.78$ (Ref. 17). This is a strong indication that the change in cot $\theta_H(T)$ is caused by the opening of the pseudogap.[20] Our result shows that, unlike the Cu NMR relaxation rate, the Zn doping does not diminish or shift the onset of the pseudogap marked by the change in cot θ_H at T_0, at least up to the Zn concentration of 1.3%. Note, however, that the deviation from the power law becomes a bit weaker (or slower) with increasing z, which is similar to what is seen in the behavior of $\rho_{ab}(T)$ (inset to Fig. 1).

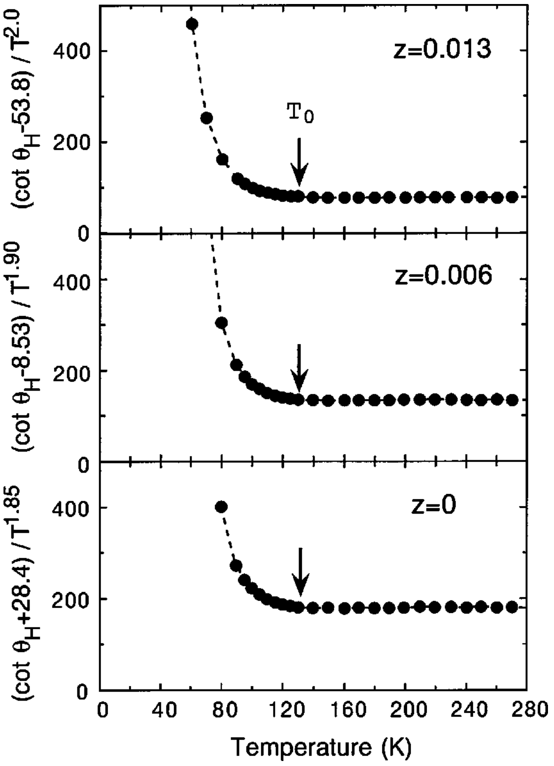

FIG. 4. Plots of $(\cot\theta_H - C)/T^{\alpha}$ vs T, which emphasizes where the deviation from the high-temperature behavior $\cot\theta_H = C + DT^{\alpha}$ (with $\alpha \simeq 2$) takes place. The deviation at T_0 is marked by arrows.

Given the fact that cot θ_H is apparently affected by the pseudogap below T_0, it is useful to clarify how the pseudogap effect is reflected in the T dependence of R_H, which is a result of the two different T dependences[3] of the more fundamental parameters τ_{tr}^{-1} and τ_H^{-1}. For this purpose, it is instructive to see how $R_H(T)$ would behave if cot θ_H continues to change as T^{α} down to T_c. The inset to Fig. 2 shows the plots of the T dependence of such hypothetical R_H^{hyp} for the three samples, where R_H^{hyp} is calculated by dividing ρ_{ab} by $(C+DT^{\alpha}) \times H$, where D is the T-independent value at temperatures above T_0 in Fig. 4. It is clear from the behavior of R_H^{hyp} that $R_H(T)$ would *not* show a peak if cot θ_H continues to change as T^{α} down to T_c. Therefore, we can conclude that the peak in $R_H(T)$ in underdoped YBCO is caused by the opening of the pseudogap.

It should be noted that the direction of the change in cot θ_H at T_0 implies that τ_H^{-1} is *enhanced* when the pseudogap opens; this is opposite to the effect on τ_{tr}^{-1}, which is *reduced* below T^*. Therefore, we cannot simply conclude that the change in cot θ_H is caused by a reduced electron-electron scattering, which is the natural consequence of a pseudogap in the low-energy electronic excitations. One possibility to understand this apparently confusing fact is to attribute the change at T_0 to the effective mass, rather than to attribute it to the scattering rate; remember that $\cot\theta_H = 1/(\omega_c \tau_H) \propto m_H/\tau_H$, where m_H is the effective mass of the particle responsible for the Hall channel,[3] so an increase in $\cot\theta_H$ is expected when the effective mass is enhanced. For example, if the pseudogap is related to the formation of dynamical charged stripes,[12] a modification in the FS topology, which leads to a change in the effective mass, is expected. This picture is also consistent with the observed robustness of the pseudogap feature in $\cot\theta_H$ upon Zn doping, because the change in the FS topology is rather insensitive to a small amount of impurities. One might question why there is little trace of the effective-mass change in the T dependence of ρ_{ab}. If $\cot\theta_H$ and ρ_{ab} reflect different parts of the FS (as is conjectured in the hot/cold spots scenario[7]), it is possible that the modification of the FS topology alters the band mass for the Hall channel while leaving that of the diagonal channel relatively unchanged.

Finally, we note that the peak in the T dependence of R_H is not always caused by the pseudogap. For example, in overdoped $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2201), it has been reported[21] that $\cot\theta_H$ shows a good T^2 dependence down to near T_c (which implies that the pseudogap does not open), and yet the peak in $R_H(T)$ is observed at a temperature well above T_c. In this case, the peak in $R_H(T)$ is just a result of the two different T dependences of $\tau_{tr}^{-1} \sim T^n + A$ $(1 \leqslant n \leqslant 1.9)$ and $\tau_H^{-1} \sim T^2 + B$ (note that in Tl-2201 both τ_{tr}^{-1} and τ_H^{-1} have somewhat large offsets even in pure crystals[21]). Mathematically, $R_H \sim (T^n + A)/(T^2 + B)$ has a peaked T dependence and thus $R_H(T)$ can show a peak well above T_c for some combination of A and B, even when both ρ_{ab} and $\cot\theta_H$ do not show any deviation from the power laws. On the other hand, as is demonstrated in the inset to Fig. 2, the peak in $R_H(T)$ of underdoped YBCO cannot be accounted for by the above origin and therefore is clearly caused by the pseudogap. This argument tells us that one should always look at the T dependence of $\cot\theta_H$, not just the peak in $R_H(T)$, to determine whether the pseudogap is showing up through $(\omega_c \tau_H)^{-1}$.

In summary, we observed that $\cot\theta_H$ of pure and Zn-doped YBCO ($y = 6.78$) crystals shows an upward deviation from the T^2 behavior below a temperature T_0 that is notably higher than T_c but is much lower than T^*. The onset temperature T_0 for this deviation, which is found to be unaffected by Zn doping, is close to the lower temperature scale for the pseudogap T_g (probed by the Cu NMR relaxation rate, for example). The fact that $\cot\theta_H$ tends to be *enhanced* below T_0 suggests that the effect of the pseudogap is *not* to reduce the Hall scattering rate; we therefore propose that the effect is more likely to be originating from a change in the Fermi surface topology, which causes a change in the effective mass. Also, we demonstrated that the peak in $R_H(T)$ of underdoped YBCO is not just a result of two different scattering times, but is actually a result of the pseudogap effect on $\cot\theta_H$.

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