The Hall effect in Zn-doped YBa$_2$Cu$_3$O$_{7-\delta}$ revisited: Hall angle and the pseudogap

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The temperature dependence of the Hall coefficient is measured with a high accuracy in a series of YBa$_2$(Cu$_{1-z}$Zn$_z$)$_3$O$_{6.78}$ crystals with 0$\leq$z$\leq$0.013. We found that the cotangent of the Hall angle, cot $\theta_H$, starts to deviate upwardly from the $T^2$ dependence below $T_0$ ($\sim$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.

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

Separately from the above development, it has become a common understanding 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 YBCO, the in-plane resistivity $\rho_{ab}$ 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. 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, 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 or charged stripe correlations; the lower temperature scale $T_g$ corresponds to the opening of a more robust pseudogap in the density of states, which can be observed by the angle-resolved photoemission or by the tunneling spectroscopy.

It was previously discussed 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, 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 $\rho_{ab}(T)$ at underdoped YBCO crystals is almost unchanged, while the suppression in the Cu NMR relaxation rate below $T_g$ is diminished with only 1% of Zn. 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$_z$)$_3$O$_{6.78}$ crystals with $y=6.78$, which corresponds to an underdoped concentration. At this composition $y=6.78$, which gives $T_c \approx 75$ 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. [14]) 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 $\cot \theta_H$ 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...
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, with 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 $\rho_{ab}$ reported here is estimated to be better than $\pm 5\%$, and the relative error in the data for each sample is less than $\pm 2\%$.

Figure 1 shows the temperature dependence of $\rho_{ab}$ for the three Zn concentrations. Above $\sim 200$ K, $\rho_{ab}$ 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 the three samples, indicating that the upper pseudogap temperature $T^*$ does not change with $z$. This result is in good agreement with the previous reports.

Figure 2 shows the temperature dependence of $R_H$ for

FIG. 1. $T$ dependence of $\rho_{ab}$ 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.

FIG. 2. $T$ dependence of $R_H$ for pure and Zn-doped samples. Inset: Plot of $R_H^{\text{hyp}}$ vs $T$ for the three samples, see text.
the three samples. Our results are somewhat different from previous results on single crystals [13], but rather resemble that of the thin film result [14]. 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 $\cot \theta_H$ is in good agreement with the previous studies; as is shown in Fig. 3, $\cot \theta_H(T)$ 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 $\tau_{tr}^{-1}$ and $\tau_H^{-1}$, 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 $R_H = \rho_{ab}/\cot \theta_H \sim \tau_H/\tau_{tr}$, $R_H$ 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, $R_H^{\text{pure}} \sim T/T^2 \sim T^{-1}$, we can infer that at high temperatures $R_H$ in Zn-doped sample should approach $R_H^{\text{pure}}$, while at low temperatures $R_H$ in Zn-doped sample is expected to become smaller than $R_H^{\text{pure}}$ (which can be easily seen when one considers $T \to 0$). 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 of the $T$ dependence of $\cot \theta_H$. 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 emphasizes where the deviation from the high-temperature behavior $\cot \theta_H = C + DT^\alpha$ (with $\alpha \approx 2$) takes place. The deviation at $T_0$ is marked by arrows.

![Figure 3](image1.png)

**FIG. 3.** Plots of $\cot \theta_H$ vs $T^2$ for the three samples.

![Figure 4](image2.png)

**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 $\alpha \approx 2$) takes place. The deviation at $T_0$ is marked by arrows.

Given the fact that $\cot \theta_H$ 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 of the more fundamental parameters $\tau_{tr}^{-1}$ and $\tau_H^{-1}$. For this purpose, it is instructive to see how $R_H(T)$ would behave if $\cot \theta_H$ continues to change as $T^\alpha$ down to $T_c$. The inset to Fig. 2 shows the plots of the $T$ dependence of such hypothetical $R_H^{\text{hyp}}$ for the three samples, where $R_H^{\text{hyp}}$ is calculated by dividing $\rho_{ab}$ by $(C + DT^\alpha) \times H$, where $D$...
is the $T$-independent value at temperatures above $T_0$ in Fig. 3. It is clear from the behavior of $R_H^{\text{pp}}$ that $R_H(T)$ would not show a peak if $\cot \theta_H$ continues to change as $T^\alpha$ 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 $\cot \theta_H$ at $T_0$ implies that $\tau_H^{-1}$ is enhanced when the pseudogap opens; this is opposite to the effect on $\tau_c^{-1}$, which is reduced below $T^*$. Therefore, we cannot simply conclude that the change in $\cot \theta_H$ 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 in Tl-2201 both $\tau_c^{-1}$ and $m_H$ is the effective mass of the particle responsible for the Hall channel $\rho_{xy}$, 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 a dynamical charged stripes [12], a modification of 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 $\rho_{xy}$. If $\cot \theta_H$ and $\rho_{xy}$ reflect different parts of the FS (as is conjectured in the hot/cold spots scenario [3]), 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$_2$Ba$_2$CuO$_{6+x}$ (Tl-2201), it has been reported [2] 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 $\tau_c^{-1} \sim T^n + A (1 \leq n \leq 1.9)$ and $\tau_H^{-1} \sim T^2 + B$ (note that in Tl-2201 both $\tau_c^{-1}$ and $\tau_H^{-1}$ have somewhat large offsets even in pure crystals [2]). 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 $\rho_{xy}$ 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 $(\omega_c T_H)^{-1}$.

In summary, we observed that $\cot \theta_H$ of pure and Zndaoped 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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