Systematic Deviation from T-Linear Behavior in the In-Plane Resistivity of $YBa_2Cu_3O_{7-y}$: Evidence for Dominant Spin Scattering

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The in-plane resistivity $\rho_{ab}(T)$ and Hall coefficient $R_H(T)$ have been measured for $YBa_2Cu_3O_{7-y}$ single crystals with various oxygen concentrations. We find that for oxygen-depleted samples $\rho_{ab}(T)$ deviates from T-linear behavior and that $R_H(T)$ deviates from $1/T$ below a temperature T_o well above T_c. The deviation coincides with the development of a gap in the spin excitations seen in recent neutron and NMR studies. This is evidence that the charge transport in the CuO_2 plane is determined by spin scattering.

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Among the striking features of high-T_c superconductors (HTSCs) are the extraordinary transport properties in the normal state [1], which are inconsistent with the conventional electron-phonon scattering mechanism. These properties have been one of the most attractive topics for investigation, since the excitation which interacts with carriers in the normal state might play an important role in superconductivity. At the hole concentration for optimum T_c, the in-plane resistivity $\rho_{ab}(T)$ is linear in temperature over a wide temperature range from just above T_c to nearly 1000 K [2, 3]. Gurvitch and Fiory [2] pointed out that the electron-phonon coupling constant λ—estimated from the absence of resistivity saturation in HTSCs—is too small to account for high T_c in terms of the electron-phonon interaction. Martin *et al.* [3] reported a T-linear resistivity down to 10 K in $Bi_2Sr_2CuO_6$. This yields an unreasonably low Debye temperature of less than 35 K in terms of the Bloch-Grüneisen formula, which represents the temperature dependence of resistivity based on the electron-phonon scattering. These facts require an alternative mechanism as the origin of the T-linear resistivity.

The most plausible and most frequently discussed candidate is scattering due to spin fluctuations in the CuO_2 plane. Nevertheless, there has been no experimental result up to now that provides convincing evidence for spin scattering. This is in part due to the absence of a detailed comparison between the transport and magnetic properties for the same system. $YBa_2Cu_3O_{7-y}$ gives a unique opportunity for a detailed comparison between charge transport and magnetism, since inelastic neutron scattering experiments [4, 5] have revealed how the spectrum of spin fluctuations evolves with varying oxygen concentration.

Here we show the results of a systematic study on the transport properties of well-characterized $YBa_2Cu_3O_{7-y}$ (Y123) single crystals with various oxygen concentrations. We have found that for reduced compounds both the in-plane resistivity $\rho_{ab}(T)$ and Hall coefficient $R_H(T)$ have characteristic temperature and oxygen-concentration dependences and that the anomalies in spin excitation (frequently referred to as "*spin gap*") observed in the normal state ($T>T_c$) [4, 5] are clearly manifest in the charge transport. We therefore conclude that spin scattering is dominant in the normal state of HTSCs.

Single crystals of $YBa_2Cu_3O_{7-y}$ were grown by a conventional self-flux method. A Y_2O_3 crucible was employed in order to avoid contamination from the crucible. Contamination is a serious problem for our purpose. In the case of Pt crucibles, for example, ~0.5% of Cu in $YBa_2Cu_3O_{7-y}$ crystal is substituted with Pt. Substituted Pt does not seriously affect the properties of the fully oxygenated compound but the effect becomes apparent in the normal-state $\rho_{ab}(T)$ as oxygen concentration is reduced, making the result irreproducible. We selected crystals with a thickness of less than 50 μm for the measurements, since transport properties are sensitive to the inclusion of flux which is often observed in thicker crystals. The oxygen concentration of the crystals was controlled by annealing them at 600 °C for 12 h in a sealed quartz tube together with ~10 g of $YBa_2Cu_3O_{7-y}$ powder which had a prescribed oxygen concentration. The oxygen concentration of annealed single crystals should be identical to that of the powder sealed with them. After annealing, the crystals in the quartz tube were slowly cooled to room temperature to avoid freezing in disorder. The annealing period is long enough for equilibration, since crystals show a very sharp superconducting transition with a T_c that is the same as that of powder sealed with them. Further annealing up to ten days made no change in transport properties. We emphasize that this careful process enables us to obtain highly reproducible, reliable, and systematic transport data even for oxygen-deficient materials, as we will show below.

Some of the crystals were detwinned by applying uniaxial stress (~10^2 kgf/cm^2) [6] at 450 °C for 40 h in a quartz tube sealed with powder as mentioned above, in order to ascertain the contribution from CuO_2 planes to $\rho_{ab}(T)$ measured for twinned crystals. The observation by the polarized optical microscope and the single crystalline x-ray diffraction confirms that the detwinning is

perfect except at the corners in the square-shaped crystal, which does not affect the measurement seriously.

Measurements of $\rho_{ab}(T)$ and $R_H(T)$ for twinned crystals were performed with a conventional configuration of 6 electrodes. Homogeneity of the crystals was checked by the coincidence of measured $\rho_{ab}(T)$ and $R_H(T)$ for both sets of voltage electrodes for each. For detwinned crystals the in-plane resistivities perpendicular and parallel to CuO chains, $\rho_a(T)$ and $\rho_b(T)$, were measured by the Montgomery method [7]. The measurements were restricted to below 300 K, since some spurious effect (rearrangement of oxygen atoms) makes $\rho_{ab}(T)$ rise steeply above 300–400 K.

Figure 1(a) shows the temperature dependence of ρ_{ab} for twinned crystals with various oxygen concentrations. With decreasing oxygen concentration ρ_{ab} increases and T_c degrades gradually. For the T_c~90 K crystal ($7-y$~6.90) the temperature dependence of ρ_{ab} is linear. That this is a property inherent to the CuO_2 planes is confirmed by the data on the detwinned crystal (shown in the inset) [8]. Although $\rho_{ab}(T)$ for slightly reduced samples shows a T-linear dependence around room temperature, it deviates from a T-linear dependence, decreasing rapidly with decreasing temperature below ~180 and ~220 K for $7-y$~6.85 and ~6.78, respectively. With further reduction, the temperature at which the deviation starts increases. This deviation of $\rho_{ab}(T)$ implies that with decreasing temperature the scattering rate of charge carriers decreases faster than that in the temperature range where $\rho_{ab} \propto T$. In order to confirm that the characteristic temperature dependence in the reduced crystals reflects the properties of the CuO_2 plane, $\rho_a(T)$ and $\rho_b(T)$ measured for a detwinned T_c~60 K crystal ($7-y$~6.68) are also shown in the inset of Fig. 1(a). The magnitude and the temperature dependence of ρ_a and ρ_b are nearly the same, implying that the anisotropy within the CuO_2 plane is very small in the oxygen-depleted material. The small anisotropy implies that the contribution of CuO chains is very small and the deviation from T linearity is inherent to the CuO_2 plane.

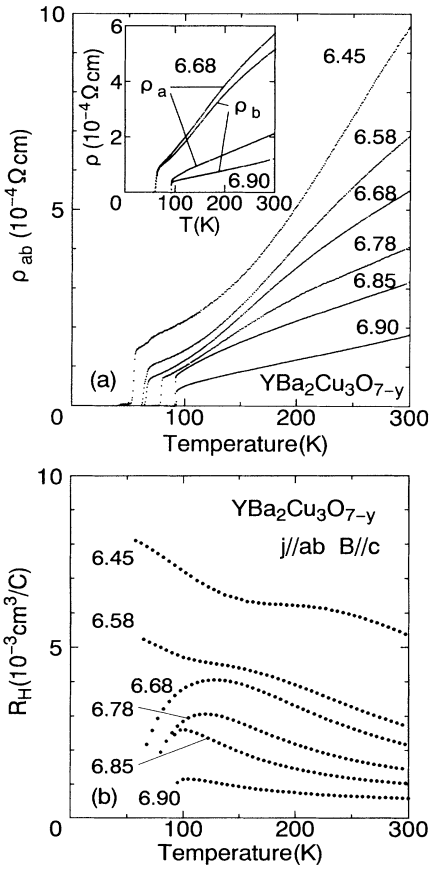

FIG. 1. (a) Temperature dependence of in-plane resistivity of twinned $YBa_2Cu_3O_{7-y}$ crystals with oxygen concentration $7-y$ ~6.90, 6.85, 6.78, 6.68, 6.58, and 6.45. Inset: Temperature dependence of ρ_a and ρ_b for detwinned crystals of T_c=90 and 60 K. (b) Temperature dependence of R_H of twinned crystals measured under $\mathbf{j} \parallel ab$ plane and $\mathbf{B} \parallel c$ axis at $B = 5$ T.

The deviation from the T-linear resistivity apparently has an intimate connection to the temperature dependence of R_H as shown in Fig. 1(b). The magnitude of R_H increases with decreasing oxygen concentration, i.e., decreasing hole concentration. Although $R_H \propto 1/T$ for fully oxygenated samples [9], for reduced samples a deviation from the $1/T$ dependence becomes apparent, with a peak occurring at a temperature well above T_c. As in the case of $\rho_{ab}(T)$, the onset of the deviation from $1/T$ increases in temperature with decreasing oxygen concentration. We find that the change of the T dependence of R_H exactly follows that of $\rho_{ab}(T)$. Note that magnetic field dependences of ρ_{ab} and R_H are not seen up to the field of 5 T used in the present experiments.

All the curves in Fig. 1(a) look very similar; ρ_{ab} is linear in T above a certain temperature T_o and shows a stronger T dependence ($\rho_{ab} \sim T^\alpha$ with α~2.5 for every y) below T_o. The crossover temperature T_o decreases with increasing oxygen concentration and approaches T_c for the 90 K crystal. It can be shown that all the resistivity curves fit on a single $\rho_{ab}(T)$ curve by scaling T with T_o and $\rho_{ab}(T)$ with $\rho_{ab}(T_o)$. This is suggestive of essentially the same scattering mechanism working in the CuO_2 plane for any hole density.

The change of $\rho_{ab}(T)$ at T_o seems reminiscent of the Bloch-Grüneisen formula, where T_o corresponds to the Debye temperature of phonons. However, the strong dependence of T_o on oxygen concentration as well as the similar change in $R_H(T)$ rules out the possibility of phonon scattering. The most plausible and perhaps the only candidate is scattering by spin fluctuations, since they have a characteristic energy of 100–1000 K and are expected to show strong dependence on the oxygen concentration.

The temperature dependence of nuclear relaxation

$(T_1T)^{-1}$ on the Cu(2) site [10, 11]—which is roughly proportional to the staggered susceptibility at $\vec{q}=\vec{Q}=(\pi,\pi)$, $\chi(Q)$, where T_1 is nuclear spin-lattice relaxation time —provides information on the spin excitation. $(T_1T)^{-1}$ for the fully oxygenated compound obeys a Curie-Weiss law. This is an indication of antiferromagnetic correlations persisting well into the superconducting regime. On the other hand, $(T_1T)^{-1}$ for the 60 K compound exhibits a deviation from Curie-Weiss law behavior at low temperatures in the normal state. The relaxation rate becomes lower below a certain temperature above T_c, giving rise to a peak in the T dependence of $(T_1T)^{-1}$. This decrease in $(T_1T)^{-1}$ is considered to originate from the reduction in the low energy spectral weight of the spin fluctuations. This has been confirmed by neutron inelastic scattering experiments [4, 5] which demonstrated the development of a *spin gap* at temperature fairly higher than the superconducting transition temperature T_c. The opening of the *spin gap* is seen at about 150 K or higher in the 60 K compounds. The temperature at which the *spin gap* becomes apparent decreases as the oxygen concentration increases and becomes indistinguishable from a superconducting gap in the 90 K materials.

The observed T and y dependences of the transport coefficients exactly follow this characteristic variation of the spin excitation spectrum, $\rho_{ab} \propto T$ and $R_H \propto 1/T$, when there is no gap in the spin excitation while a deviation from these dependences is seen upon opening of the *spin gap*.

In order to get insight more into the charge dynamics and to emphasize the deviation from T-linear resistivity, we replot the data as $[\rho_{ab}(T)-\rho_{ab}(0)]/\alpha T$ vs T in Fig. 2. Here, $\rho_{ab}(0)$ is the T=0 intercept of the line extrapolated from the T-linear part and α is the slope of the T-linear resistivity. It seems that the data in Fig. 2 mimic $\chi(0)$, as determined by the NMR Knight shift. The NMR Knight shift at the ^{89}Y site [12]—which is proportional to the uniform susceptibility at q=0, $\chi(0)$—is almost temperature independent for fully oxygenated compounds. When the compound is reduced, $\chi(0)$ decreases with lowering temperature.

The decrease of $\chi(0)$ is interpreted in the theories based on the resonating valence bond (RVB) picture as arising from the *spin gap* due to the formation of spin singlets [13] and is linked to the behavior of $\chi(Q)$ [14]. The temperature below which the spin singlets are formed increases with reducing hole density as the observed T_o does. In the gauge field theory of the uniform RVB state [15] the charges are scattered by the fluctuating gauge fields which are related to the fluctuations in the spin chirality. The fluctuations are suppressed by the formation of spin singlets, leading to the reduction in resistivity. The similarity between $\chi(0)$ and the plots in Fig. 2 is suggestive of this mechanism. Since the energy for spin singlet or RVB formation is the order of the superexchange coupling J ($\sim$ 1000 K), ordinary magnetic fields (10 T or lower) are too weak to affect the singlet formation, consistent with the negligibly small effect of magnetic field on $\rho_{ab}(T)$ and $R_H(T)$ below T_o.

There is another school of theorists that explains the T-linear dependence of ρ_{ab} in terms of spin scattering using Fermi liquid theory. In these theories, the two-dimensional electrons are scattered by antiferromagnetic spin fluctuations [16, 17]. According to the self-consistent renormalization theory for spin fluctuations [16, 18], ρ_{ab} is proportional to $\sim\chi(Q)T^2$. We show in Fig. 3 a plot of $[\rho_{ab}(T)-\rho_{ab}(0)]/T^2$ vs T for two representative compositions. The similarity between the data in Fig. 3 and NMR $(T_1T)^{-1}$ [10, 11] might be suggestive of this mechanism. However, it is not clear if the presence of a *spin gap* as well as the temperature-dependent R_H are compatible with the Fermi liquid picture.

In Fig. 2 the resistivities are normalized by the slope α of the T-linear part of ρ_{ab}. The value of α^{-1} is plotted in the inset as a function of oxygen concentration.

In the same figure the Drude spectral weight ($\omega_{pD}^2 \sim n/m^*$, n and m^* being carrier density and effec-

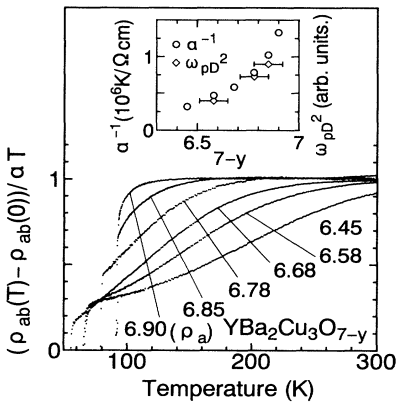

FIG. 2. $[\rho_{ab}(T)-\rho_{ab}(0)]/\alpha T$ as a function of temperature, where α is the slope of the T-linear region of ρ_{ab} (ρ_a) curve and $\rho_{ab}(0)$ is the extrapolated value to T=0 K from the T-linear region. Inset: Oxygen-concentration dependence of α^{-1} and the Drude spectral weight (ω_{pD}^2) [19].

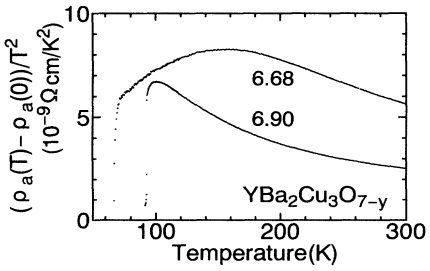

FIG. 3. $[\rho_a(T)-\rho_a(0)]/T^2$ vs T for 60 and 90 K crystals, ρ_a being the T=0 K intercept of the T-linear part. The T dependence of the data is similar to that of the NMR T_1T data [11].

tive mass, respectively) is also plotted. These values were taken from Table I in Ref. [19], which estimated the Drude spectral weight from the magnetic penetration depth or the optical conductivity spectrum. Both α^{-1} and ω_{pD}^2 show the same y dependence. Considering that $\rho_{ab}=(4\pi/\omega_{pD}^2)\tau^{-1}$, where τ^{-1} is the scattering rate and $\tau^{-1}=2\pi\lambda T$ in the T-linear region, the result indicates (1) that the variation of ρ_{ab} with y is caused primarily by the change of ω_{pD} and (2) that the carrier scattering rate τ^{-1} is nearly the same in the T-linear region for any oxygen concentration. This is equivalent to saying that the coupling strength λ between the charge carriers and some excitations (spin excitations) does not appreciably change with y. We calculate $\lambda\sim0.3$ from the measured ρ_{ab} (ρ_a) values. This is in agreement with the estimate of τ from the width of the Drude optical spectrum [19].

Note that ω_{pD} here is estimated by decomposing the optical conductivity into Drude and so-called mid-infrared parts. There is an alternative interpretation of the optical spectrum where the optical conductivity is expressed by a single Drude formula assuming ω- (and T-) dependent τ. Such an analysis on the optical spectra of $YBa_2Cu_3O_{7-y}$ suggested that τ^{-1} might be linear both in T and ω, $\tau^{-1}=2\lambda\max(\pi T,\omega)$ [20] with λ increasing with reducing oxygen concentration. In this case one would expect that the ω dependence of τ might change below a certain frequency ω_o ($\sim\pi T_o$). However, it is difficult to see such a crossover in the available optical data for 60 K $YBa_2Cu_3O_{7-y}$ [20,21] because of relatively large uncertainty in such a low ω region.

The deviation from T-linear dependence of ρ_{ab} has also been observed in $YBa_2Cu_4O_8$ (Y124) [22] and lightly doped $La_{2-x}Sr_xCuO_4$ (La214) [23]. The behavior of ρ_{ab} of Y124 [22] appears essentially the same as that of reduced $YBa_2Cu_3O_{7-y}$ (Y123). Y124 is expected to have the same spin-fluctuation spectrum as that seen in Y123 in the underdoped region. In the La214 system [23], the deviation appears to occur at nearly the same temperature for every Sr composition. A possible reason for the difference might be the different spin excitation spectrum. Instead of a *spin gap* for Y123 [4, 5], an incommensurate modulation is observed for La214 [24].

In summary, we have clearly established an intriguing correlation between the deviation of the in-plane resistivity from T linearity and the change in the spectrum of spin fluctuations.

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