Extraordinary Hall effect in $YBa_2Cu_3O_{7-\delta}$ superconductors

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The paramagneticlike T^{-1} temperature dependence of the Hall coefficient R_H and the linear-T resistivity ρ are shown to be obeyed in the oxygen-deficient metallic phases of the $YBa_2Cu_3O_{7-\delta}$ superconductor at elevated temperatures, a region of oxygen stoichiometry $0.03 \lesssim \delta(T) \lesssim 0.6$, as revealed by a universal $R_H T$ vs ρ/T dependence. The similarly temperature-scaled Hall angle increases upon oxygen desorption and has a maximum near $\delta \approx 0.2$. These results suggest that asymmetric magnetic scattering may contribute significantly to the Hall effect in $YBa_2Cu_3O_7$.

The key motivation for Hall-effect studies in the high-T_c superconductors has been to ascertain the sign and concentration of the carriers.[1–6] For transport along the CuO_2 planes of $YBa_2Cu_3O_7$, the Hall coefficient $R_H(T)$ has a puzzling paramagneticlike T^{-1} temperature dependence,[1,2] making the interpretation model dependent [e.g., ~ 1 hole per Cu (Ref. 6)]. The in-plane resistivity $\rho(T)$ of $YBa_2Cu_3O_7$ is linear in T up to at least ~ 800 K, as shown by single-crystal work.[7] In this report, we demonstrate that these temperature laws generally extend into the oxygen-deficient phases of $YBa_2Cu_3O_{7-\delta}$ at high temperatures, a result which is not obvious from inspection of $\rho(T)$ and $R_H(T)$ curves themselves because of the strong temperature dependence of $\delta(P_{O_2},T)$.[6,8] It is revealed through the universal relationship which is displayed by the temperature-scaled quantities $R_H T$ and ρ/T at temperatures high enough to form a homogeneous oxygen-defect system which is in equilibrium with the external O_2 atmosphere.

In $YBa_2Cu_3O_{7-\delta}$ the stoiciometry is determined by occupancy of oxygen sites in the CuO chain structure between the CuO_2 double planes.[9] Ordering of the oxygen vacancies in oxygen-deficient material leads to an intricate δ-T phase diagram and superlattice structures,[10] so that an important experimental question we must address first is whether the oxygen vacancy distribution of a given sample is uniform. Figure 1 illustrates the equilibrium issue quite vividly, in a plot of the Hall coefficient in a polycrystalline $YBa_2Cu_3O_{7-\delta}$ ceramic above room temperature at various oxygen partial pressures. Both R_H and resistivity ρ generally increase with δ, as oxygen is lost at elevated temperature and reduced P_{O_2}, and this effect competes with the respective T^{-1} and T behaviors. The temperature dependences of the R_H curves display distinct minima at kinetic barrier temperatures T_K, which vary from 280 °C at $P_{O_2}=1$ atm to 400 °C at 5×10^{-5} atm. Data in the $T>T_K$ region are reversible, in the sense that equilibrium may be reached after 12–24 h, which is sufficient for oxygen to diffuse within the ceramic grains. The relaxation time was determined by monitoring the resistivity.[11] The $R_H(T)$ points taken after cooling below T_K are also reversible if T_K is not breached. However, the $T<T_K$ region is not in equilibrium with P_{O_2} and, thus, in general there is little assurance that oxygenation is homogeneous. Kinetic barriers cause $\rho(T)$ and T_c to be sensitive to oxygen cycle history. Measurements of R_H and ρ in polycrystals are converted to corresponding a-b plane values by multiplicative correction factors,[6,12] $1.25f$ and $0.5f$, respectively, where f is a filling factor (0.95 in the present case). The results for $\delta \approx 0$ agree with data on single crystals.[3]

Figure 2 presents a plot of $R_H T$ vs ρ/T which demonstrates the aforementioned temperature scaling of the Hall effect and resistivity at high temperatures. The □ symbols denote points above T_K and the ● symbols below T_K, with variation according to $\delta(P_{O_2},T)$ being implicit. The oxygen stoichiometry variation in these data is $0.02 \le \delta \le 0.6$, over which range $YBa_2Cu_3O_{7-\delta}$ remains metallic and superconducting.[4,5] Generally, the plotted quantities increase with δ, or the fraction of oxygen vacancies.[6] The universal relationship among the equilibrium $T>T_K$ data points is revealed by such a plot; the spline curve is a fit to the □ points. If the T scaling were to be ignored, one obtains a series of nonoverlapping ρ-R_H curves for the various isobars, rather than a universal function. The ● points for $T<T_K$ which lie off the

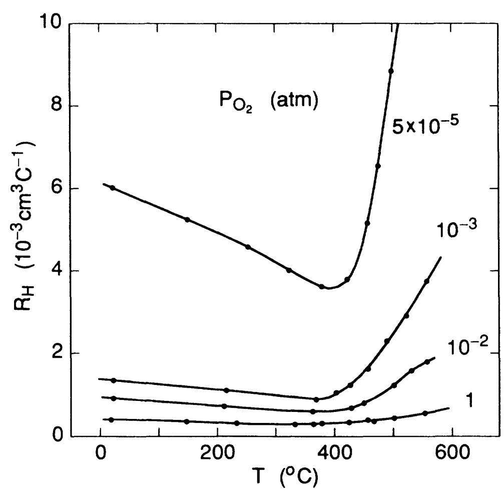

FIG. 1. Temperature dependence of the Hall coefficient in polycrystalline $YBa_2Cu_3O_{7-\delta}$ at various oxygen gas partial pressures P_{O_2}. The oxygen-exchange kinetic barrier temperature T_K for each P_{O_2} curve is at the minimum. Points for $T<T_K$ were taken after cooling slowly through T_K.

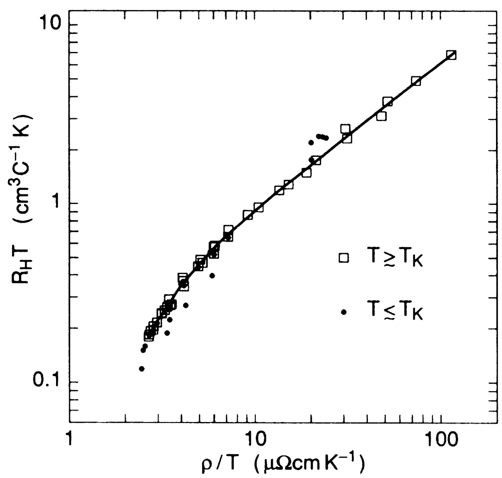

FIG. 2. Hall coefficient of $YBa_2Cu_3O_{7-\delta}$ scaled by T is plotted against resistivity scaled by T^{-1}, with (P_{O_2},T) implicit variables. Points are distinguished with respect to kinetic temperatures T_K. The solid curve follows the universal behavior of the equilibrium (□) points. The data lie in the metallic phase, $0.02 \leq \delta \leq 0.6$.

universal curve may indicate lack of equilibrium.

The increase in ρ/T and $R_H T$ with oxygen desorption is the result qualitatively expected for the decrease in carrier density. However, the behavior of the Hall angle or the Hall mobility is anomalous, since oxygen desorption and disorder do not lead to a simple monotonic decrease. Figure 3 shows this through a plot of the temperature-scaled Hall mobility (Hall angle per unit magnetic field) $\mu_H T^2$ against scaled Hall coefficient. Only equilibrium points for $T > T_K$ are plotted. We observe that the Hall angle initially *increases* by 50% upon removing oxygen, in a region that corresponds to $\delta \lesssim 0.2$. Elastic scattering associated with disorder takes over at larger δ, acts to reduce μ_H, and gives a maximum in the curve.

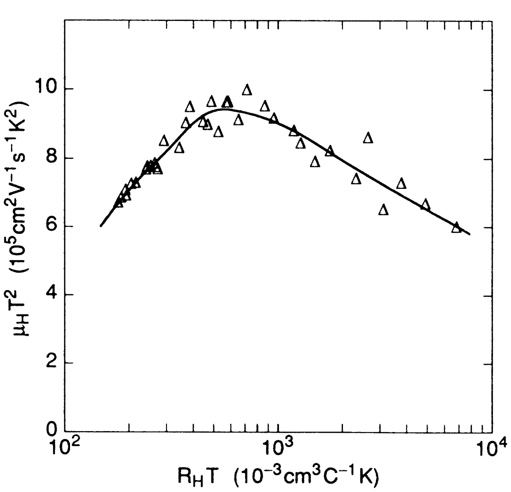

FIG. 3. Hall mobility scaled by T^2 against Hall coefficient scaled by T. Points are data for $T > T_K$; curve is a spline fit. The initial rise of 50% corresponds to the region $\delta \lesssim 0.2$.

The initial increase in the Hall angle with δ and the dominant T^{-1} temperature dependence of R_H together indicate an extraordinary Hall effect, and, thus, a magnetic skew scattering mechanism ought to be seriously considered as an explanation. The variation in the Hall coefficient with δ in $YBa_2Cu_3O_{7-\delta}$ (Refs. 4–6) shows that qualitatively the density of holes is controlled by chemical doping with oxygen. At sufficiently low carrier density, the superconductors transform into antiferromagnetic insulators and an intermediate spin-glass phase has been postulated.[13] Although a search for magnetic modes in $YBa_2Cu_3O_7$ by neutron scattering initially gave null results,[14] recent Raman scattering spectra show a broadened peak which we may presume to be damped magnetic fluctuations.[15] Magnetic fluctuations may also explain the large non-Korringa relaxation for Cu nuclei in the CuO_2 planes.[16] Oxygen-deficient material has a paramagnetic susceptibility which increases with δ,[4] further evidence for fluctuating local moments. The implication of these results is that the magnetic fluctuation spectrum is strongly weighted towards zone-boundary wave vectors.

Because of the layered $YBa_2Cu_3O_7$ structure, carriers in the CuO_2 planes are spatially separated from the moments, if the latter are located predominantly in the CuO chain structure. For detecting these magnetic fluctuations by transport measurements, rapid relaxation and short correlation lengths are probably not as relevant as in spectroscopic studies, owing to the much shorter time scales associated with electron scattering events. We therefore propose interpreting the T^{-1} dependence of R_H as an extraordinary contribution to the Hall coefficient from magnetic scattering,[17] which adds to the ordinary Lorentz force $1/nq$ term. The argument is based on the remarkable similarity between results for this superconductor and alloys containing dilute concentrations of transition-metal or rare-earth impurities (e.g., Cu:Fe, Au:Fe, and Al:Gd),[18,19] paramagnetic glasses,[20] mixed-valence and heavy-fermion alloys (e.g., $CeCu_2Si_2$).[21] In the classical description, paramagnetic moments of concentration c add a positive term in the Hall angle proportional to cH/T so that the temperature dependence of R_H reflects that of the susceptibility. A number of basic mechanisms have been invoked, essentially side jump and skew scattering caused by orbital interaction with the local moment.[17–18,21] Because consideration of skew scattering in the layered $YBa_2Cu_3O_{7-\delta}$ cuprate involves dealing with a significantly different crystal structure, a new theory will be needed.

The anomalous Hall effect should saturate at strong magnetic fields.[17–19] A dependence on magnetic field has not been reported, however, possibly because the high T_c of $YBa_2Cu_3O_7$ and the interference with superconducting fluctuations precludes one from probing the regime at high H/T. As T_c is approached, the T^{-1} divergence in R_H recovers in a narrow region and displays a sharp peak,[3] evidently as superconductive pairing quenches the paramagnetic normal-state behavior. From superconducting penetration depth experiments, which give essentially n/m^*,[22] it is certain that the carrier density does not freeze out linearly with T at low temperature. In our analysis, the ordinary Lorentz-force term in R_H has not been separated because the temperature-scaling law implies it can be neglected. From the maximum scatter of the points in Fig. 2, we estimate that the ordinary term is $\lesssim 20\%$ of R_H.

The T^{-1} law for the Hall coefficient and the linear-T law for ρ is shown to hold in the region of δ-T phase diagram of $YBa_2Cu_3O_{7-\delta}$, where the oxygen vacancies are in equilibrium with an external O_2 gas pressure. In the range $0.03 \lesssim \delta \lesssim 0.2$, oxygen desorption causes the Hall angle to increase by up to 50%. Based on striking similarity to the extraordinary Hall effect caused by asymmetric scattering in metallic alloys containing paramagnetic impurities, it is anticipated that a very similar mechanism of scattering by magnetic fluctuations occurs in the metallic-superconducting phases of $YBa_2Cu_3O_{7-\delta}$, including $\delta = 0$.

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