Variable-range hopping and positive magnetoresistance in insulating $Y_{1-x}Pr_xBa_2Cu_3O_7$ crystals

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We have measured in-plane resistivity and magnetoresistance of insulating $Y_{1-x}Pr_xBa_2Cu_3O_7$ ($x\sim0.63$) crystals in magnetic fields up to 8 T. Mott variable-range-hopping transport $\rho=\rho_0\exp\{(T_0/T)^\alpha\}$ has been observed with $\alpha=\frac{1}{4}$ at low temperatures suggesting the metal-insulator transition caused by Pr doping is due to carrier localization. The magnetoresistance in the Mott variable-range-hopping regime is found to be positive and anisotropic, which may be caused by a decrease of the localiztion length induced by the magnetic field.

It is well known that substituting Y by Pr gradually drives $YBa_2Cu_3O_7$ (YBCO) from a high-T_c superconductor ($T_c\sim90$ K) to an insulator with a metal-insulator transition (M-I) occurring near Pr doping ~0.55, in sharp contrast to other rare-earth substitutions which show little effect on T_C. The important role played by Pr doping in suppressing the superconductivity and causing the M-I transition has received much attention but it has not yet been fully understood.[1] While a model has been proposed in which Pr has a valence state greater than 3+ and thus fills holes in CuO_2 planes, high-energy spectroscopy data[2] strongly suggested trivalent Pr and thus support a localization picture based on the concept of strong hybridization between Pr^{3+} $4f$ states and Cu-O valence-band states.[3] In a previous study,[4] we proposed that Pr dopants introduce a disordered potential into the CuO_2 planes which may localize electrons and contribute to the suppression of superconductivity. It is thus of interest to study the transport properties of $Y_{1-x}Pr_xBa_2Cu_3O_7$ on the insulating side which may shed light on the nature of the M-I transition induced by Pr doping.

In systems with localized electronic states, it is often observed that variable-range-hopping (VRH) conduction occurs at low temperatures.[5] VRH has been observed in some insulating high-T_c cuprates, such as $La_{2-x}Sr_xCuO_4$,[6,7] $PrBa_2Cu_3O_7$,[8] $YBa_2Cu_{3-x}(Fe,Zn)_xO_7$, and $YBa_2Cu_3O_6$.[9] These observations suggest that the M-I transitions in these systems are caused by the localization of the charge carriers rather than by the opening of an energy gap at the Fermi level. In some cases,[8] however, it was found to be difficult to determine the dimensionality of the VRH which may be partially due to the polycrystalline nature of the ceramic samples used. In order to eliminate the grain-boundary effects and study the intrinsic electronic properties, it is necessary to use single-crystal samples. In this work, we report a transport study of barely insulating $Y_{0.37}Pr_{0.63}Ba_2Cu_3O_{7-\delta}$ (YPBCO) crystals which show the Mott VRH conduction at low temperatures, suggesting that the M-I transition induced by Pr doping in YPBCO is also caused by a strong disorder. By measuring the anisotropy of the magnetoresistance, we have further studied an interesting phenomenon, i.e., the influence of a magnetic field on the transport of localized electrons in the VRH regime which has recently attracted considerable attention in strongly disordered materials.[10,11]

YPBCO crystals were grown by a self-flux method.[4] To get fully oxygenated samples, we annealed the crystals in flowing oxygen at about 450 °C for one week. EDX (energy dispersive x-ray) measurements done on one of the two samples used in this study (sample A) shows that the Pr content is 0.63 ± 0.01 and uniform. We assume the oxygen content is close to O_7 after the long-time oxygen annealing. A four-probe method is employed on rectangular-shape crystals to measure the in-plane resistivity. Low contact resistance of a few ohms is achieved by soldering thin gold wires with indium-silver alloy onto the crystals. The samples were mounted on a rotatable stage (a Hall sensor is fixed to this stage) in a superconducting solenoid so that the anisotropy of the magnetoresistance can be measured. Using carbon-gas resistor thermometry (compensated for the small magnetoresistance of the sensor at high fields and low temperatures[12]), we achieved temperature stability better than ±20 mK as the magnetic field was varied.

The in-plane resistivity ρ of our YPBCO samples is shown in the inset to Fig. 1 as a function of T. The insulating behavior has been verified by plotting the conductivity ($=1/\rho$) vs T which extrapolates to zero at $T=0$. Using the resistivity value at the room temperature, the sheet resistance for a CuO_2 layer (defined as ρ/d, $d\approx12$ Å is the interlayer spacing) is calculated to be $\sim7000\Omega$, which is above the critical value $h/4e^2$ for a localization-induced superconductor-insulator transition in a two-dimensional (2D) system.[13] In strongly disordered materials, VRH transport is usually expected to occur and the resistivity is given by the Mott formula $\rho=\rho_0\exp(T_0/T)^\alpha$ with $\alpha=1/(d+1)$ and d the dimensionality of the system, assuming a constant density of states (DOS) $N(\epsilon_F)$ at the Fermi level.[5] The parameter T_0 is related to $N(\epsilon_F)$ and the localization length ξ as $kT_0\approx21/[N(\epsilon_F)\xi^3]$ in 3D. The temperature dependence of ρ_0 is generally weak and can be neglected. Shown in Fig. 1 is the semilog plot of ρ vs $T^{-1/4}$ for both our samples over the temperature range $1.5\leq T\leq300$ K. A linear variation is clearly observed below a characteris-

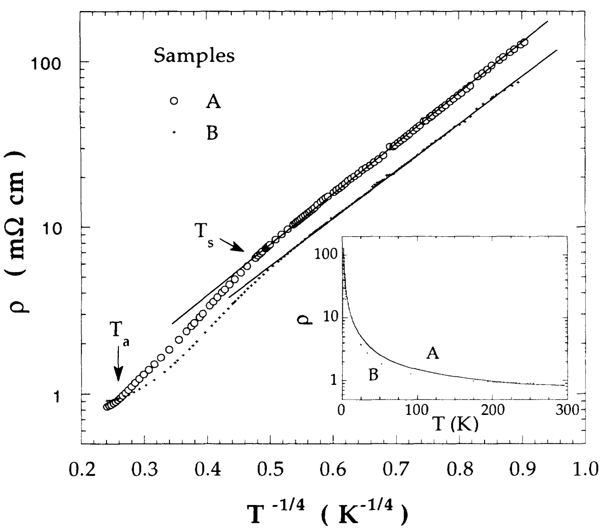

FIG. 1. Semilog plot of ρ vs $T^{-1/4}$ for $Y_{0.37}Pr_{0.63}Ba_2Cu_3O_7$ crystals. The solid lines are the best linear fit to the low-T data. Inset: lnρ vs T.

tic temperature T_a ($\sim$220 K for sample A and $\sim$100 K for sample B) suggestive of VRH transport at low temperatures. This is consistent with earlier thermopower data in sintered samples which implies no energy gap near the Fermi surface.[14] A possible crossover from the VRH to nearest-neighbor hopping occurs at $\sim T_a$. The difference in ρ between the two samples could be due to a slightly different Pr or oxygen content. One noticeable feature is the slope change at $T_s \sim 20$ K which is nearly coincident with the Néel temperature T_N ($\sim$30 K) for the AFM ordering of the planar Cu^{2+} spins in Pr-doped YBCO ($x \sim 0.6$) as observed by muon spin relaxation (μSR).[15] As the Cu moments order, the resistivity is reduced because spin scattering of the carriers is greatly suppressed.

For T above 20 K, we cannot definitely determine whether $\alpha = \frac{1}{3}$ or $\frac{1}{4}$ is more valid due to the limited temperature range. For $T \leq 16$ K, however, a value of $\alpha = \frac{1}{4}$ seems to fit the data better than $\alpha = \frac{1}{3}$, although our data spans only a decade in the resistivity value. Our low-T magnetoresistance (MR) measurements also suggest 3D-like VRH. At $T \leq 8$ K, a large positive MR was observed when H is applied in the CuO_2 planes and $\Delta R(H)/R(0)$ has almost the same magnetic field dependence as that of the MR with H parallel to the c axis (except for a scaling factor), as shown in the inset to Fig. 3(b) for $T = 2.1$ K. It is thus likely that part of the MR for both field orientations is caused by a similar scattering mechanism, suggesting a possible anisotropic 3D-like conduction. This might result from the strong hybridization between the Pr 4f wave function and the conduction-band orbitals of the CuO_2 planes. Racah, Dai, and Deutscher[16] also suggested a possible 3D-like transport in Pr-doped YBCO based on their studies of the superconducting transition under applied magnetic fields in YPBCO films with $x \approx 0.4$. We emphasize, however, that our present work by itself is not sufficient to draw any definitive conclusion about the dimensionality of the charge transport in YPBCO. More measurements at much lower temperatures are certainly needed.

If we assume that a 3D-like VRH occurs in these YPBCO crystals, the parameter T_0 in the Mott formula can be extracted from the low T data, and we find 2420 and 2163 K for samples A and B, respectively. Assuming $N(\epsilon_F) \sim 10$ states/eV cell based on specific-heat measurements[17] on YPBCO, the localization length ξ estimated from T_0 is about 12–15 Å which is about three in-plane lattice spacings. The most probable hopping distance $R \approx 0.4\xi(T_0/T)^{1/4}$ is about 16–20 Å at 20 K so $R/\xi \geq 1.4$ for $T \leq 20$ K, fulfilling the requirement for VRH to be appropriate.

In order to see if the VRH conduction correlates with Pr doping, we plot the resistivity of a $PrBa_2Cu_3O_7$ (PBCO) crystal from our previous work[4] in Fig. 2 as lnρ vs $T^{-\beta}$. Though the temperature range is limited ($120 \leq T \leq 300$ K), we find that neither 2D ($\beta = \frac{1}{3}$) nor 3D ($\beta = \frac{1}{4}$) VRH can fit the data well. Instead, $\beta = \frac{1}{2}$ gives a much better fit, suggesting the presence of a Coulomb gap in the DOS at the Fermi energy.[5] This result differs from that of Fisher *et al.* on PBCO ceramics and films.[8] They found a β close to $\frac{1}{3}$ or $\frac{1}{4}$. We tentatively attribute this difference to the sample variations. It is possible, however, that β may change at low temperatures. The $\beta = \frac{1}{2}$ behavior has also been seen[6] in strongly insulating samples of $La_{2-x}Sr_xCuO_4$ at low T. As the M-I transition is approached from the insulating side, β generally changes to $\frac{1}{4}$ for a 3D system since the Coulomb interaction is screened by the increase of the carrier density.

In the following, we will focus on the MR data at T below $\sim$16 K. The field dependence of the magnetoresistance $\Delta R(H)/R(0)$ of sample A at low T is shown in Fig. 3 for both field orientations. Sample B has a very similar

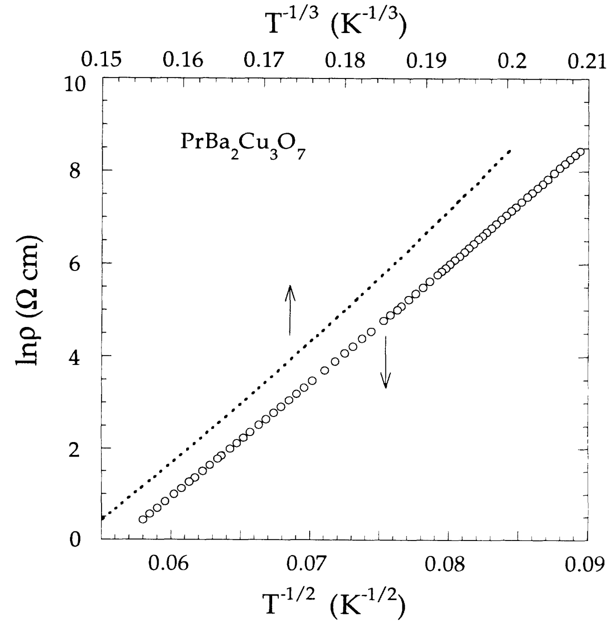

FIG. 2. lnρ vs $T^{-\beta}$ for a $PrBa_2Cu_3O_7$ crystal.

behavior and is therefore not shown. We find that the MR is positive below ~ 16 K for both field directions and gets larger as T decreases. Above ~ 16 K, the perpendicular MR becomes small and negative which may be caused by the suppression of the scattering of the electrons from the Cu spins at $T \geq T_N$ since the spins are partially aligned by the field. The magnetoresistance is anisotropic, being about 22% at 2.1 K and 8 T for $H \| c$ and only 12.5% at the same temperature and field for $H \| a\text{-}b$ plane, but the field dependence is very similar for both H orientations at $T \leq 8$ K. At low fields, $\ln[R(H)/R(0)]$ is proportional to H^2 and then gradually changes to an $\sim H^{2/3}$ dependence at high fields. The difference in $\Delta R(H)/R(0)$ between $H \| c$ and $H \| a\text{-}b$ decreases monotonically as T increases.

Very recently, Iwasaki *et al.*[17] have reported magnetoresistance measurements in magnetic fields up to 14 T on $Y_{1-x}Pr_xBa_2Cu_3O_7$ films with $x \geq 0.6$. Our data are very similar to that of their $x=0.7$ sample. The MR of the $x=0.7$ film was found to be anisotropic and positive at low T (≤ 10 K) and tends to be negative at a temperature around 16 K. The field dependence of $\Delta R/R$ and the temperature dependence of its anisotropy are fully consistent with our observations. The magnitude of MR of the thin film is larger than that of our crystals which could be due to a higher Pr content in their film sample. For other Pr-doped films, they found that MR is always positive for $x=1$ but exhibits a more complicated behavior for the $x=0.6$ film: the MR is positive only at low T and low fields and then becomes negative at high fields and high temperatures. Based on these results, they proposed that there are at least two kinds of scattering mechanisms contributing to the observed anomalous MR, although they could not identify these scattering terms. Our present single-crystal work also reveals the existence of both positive and negative terms in the magnetoresistance of YPBCO.

Positive magnetoresistance in the VRH regime has been observed in many strongly disordered materials. It has been interpreted[11,19] as the result of a decrease of the localization length induced by the magnetic field. While it is not clear if these models are valid in strongly correlated, layered systems like high-T_c oxides, it is instructive to see if such a mechanism can possibly account for the positive MR in YPBCO. For magnetic fields up to 8 T, we find that $\rho(H)$ vs T of YPBCO crystals continues to fit the VRH behavior with the slope ($\propto T_0$) increasing as H increases as shown in Fig. 4, assuming a 3D-like VRH. Since $T_0 \propto 1/\xi^3$, the increase of T_0 indicates the decrease of the localization length $\xi(H)$ in a magnetic field if $N(\epsilon_F)$ is not field dependent. In the inset to Fig. 4, we plot $\xi(H)/\xi(0)$, calculated from the slope of $\ln\rho(H)$ vs $T^{-1/4}$, as a function of H which clearly shows that $\xi(H)$ decreases with increasing H and tends to saturate at high fields (> 10 T). This behavior of ξ is qualitatively the same if we assume a 2D-like VRH in YPBCO.

In conclusion, we have measured in-plane resistivity and magnetoresistance in barely insulating $Y_{0.37}Pr_{0.63}Ba_2Cu_3O_7$ crystals. We find that the resistivity follows the Mott VRH conduction $\rho \approx \rho_0 \exp\{(T_0/T)^\alpha\}$ with a change in T_0 at about 20 K which is very likely to

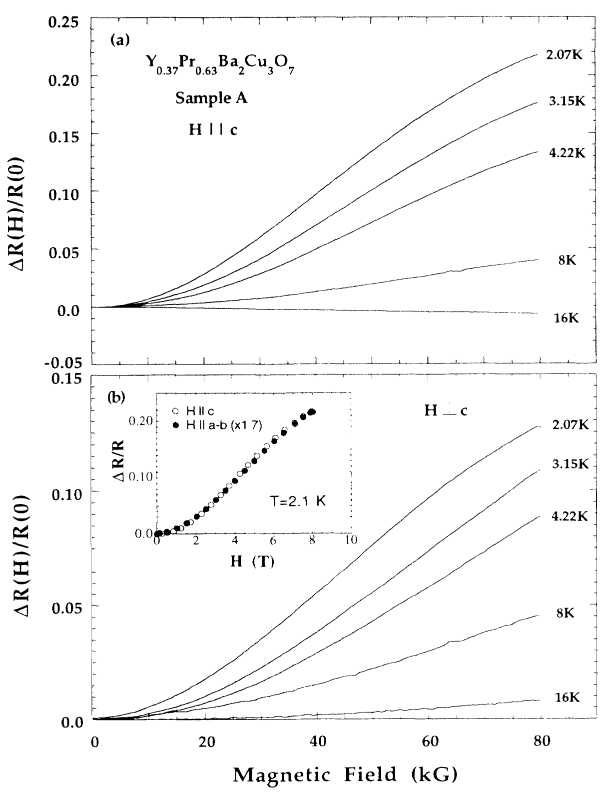

FIG. 3. The magnetoresistance of sample A as a function of H at various temperatures with (a) H parallel to the c axis and (b) H parallel to the CuO_2 planes. The inset shows the field dependence of $\Delta R/R$ for both H directions at $T=2.1$ K.

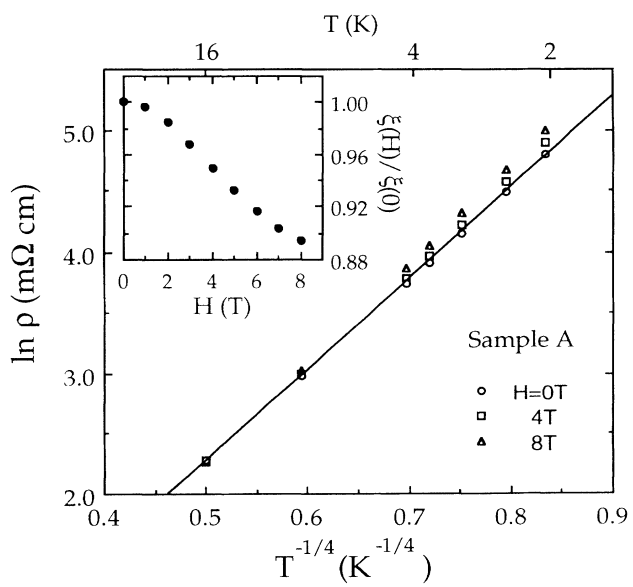

FIG. 4. The plot of $\ln\rho$ vs $T^{-1/4}$ for sample A at $H=0$, 4, and 8 T. The solid line is the best linear fit to the $H=0$ data. The inset shows the variation of the localization length ξ with magnetic field.

be caused by the AFM ordering of the Cu^{2+} spins in the CuO_2 planes. At T below ~ 16 K, we have observed an anisotropic, positive magnetoresistance which might be due to a magnetic field induced decrease of the localization length as seen in some strongly disordered semiconductors. Our results suggest that the metal-insulator transition induced by Pr doping in YPBCO is caused by the localization of the charge carriers.

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