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Magnetic ordering and chain transport of $PrBa_2Cu_3O_{7-y}$

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The effects of Fe, Co, and Ni substitution on the structural, magnetic, and electrical properties of $PrBa_2Cu_3O_{7-y}$ have been investigated by means of powder x-ray diffraction, magnetization, and electrical-resistivity measurements. All three systems $PrBa_2(Cu_{1-x}M_x)_3O_{7-y}$ (M = Fe, Co, and Ni) exhibit an orthorhombic-tetragonal transition at about $x=0.025$ for Fe and Co, and $x=0.05$ for Ni. The magnetic ordering temperature T_N of $PrBa_2Cu_3O_{7-y}$ is reduced by the doping of Fe, Co, and Ni with the depression rate $dT_N/dx \sim 0.65$, 0.75, and 0.45 K/at. %, respectively. The doping of Fe or Co has a larger effect than Ni on the increase of room-temperature resistivity. Combining these results with those observed previously in Zn- and Ga-doped $PrBa_2Cu_3O_{7-y}$, it is interpreted that the Cu-O chain is considered to play a crucial role in the magnetic-coupling mechanism of $PrBa_2Cu_3O_{7-y}$.

I. INTRODUCTION

The quenching of superconductivity and the anomalous magnetic properties of $PrBa_2Cu_3O_{7-y}$ have been received much attention since the material was synthesized. $PrBa_2Cu_3O_{7-y}$ forms the same orthorhombic structure as $YBa_2Cu_3O_{7-y}$ and other rare-earth high-T_c compounds, but it is strangely not superconducting.[1] A number of experimental results and the mechanisms proposed for the depression of superconductivity in $Y_{1-x}Pr_xBa_2Cu_3O_{7-y}$ were reviewed in a series of recent papers (see Refs. 2–12, and the references quoted therein). To our knowledge, the nature of the quenching of superconductivity in $PrBa_2Cu_3O_{7-y}$ is not yet clear. Recently, Norton *et al.*[13] reported the superconductivity with $T_c \sim 40$ K in $Pr_{0.5}Ca_{0.5}Ba_2Cu_3O_{7-y}$ thin film. This results strongly supported the hole localization and filling mechanism for the suppression of superconductivity by Pr in $YBa_2Cu_3O_{7-y}$. In the meanwhile, an antiferromagnetic order in $PrBa_2Cu_3O_{7-y}$ was observed below $T_N=17$ K by magnetic susceptibility and heat capacity.[7,9] Neutron-diffraction line shapes and intensity indicated that the ordering was of Pr moments and the superexchange mechanism involving Cu-O_2 planes was dominant in determining Pr magnetism.[14] If it is Pr that orders, this magnetic ordering temperature 17 K of $PrBa_2Cu_3O_{7-y}$ is about 2 orders-of-magnitude higher than expected if one scales the T_N for $GdBa_2Cu_3O_{7-y}$ ($T_N=2.2$ K) assuming either purely dipolar interactions or Ruderman-Kittel-Kasuya-Yosida (RKKY) exchange. Therefore it is interesting to understand the correlation between the quenching of superconductivity and the unusual magnetic properties of $PrBa_2Cu_3O_{7-y}$. Furthermore, the magnetic coupling mechanism in $PrBa_2Cu_3O_{7-y}$, whether it is identical to that in $GdBa_2Cu_3O_{7-y}$ or not, is also intriguing.

The magnetic ordering temperature T_N in $Gd_{1-x}Pr_xBa_2Cu_3O_{7-y}$ decreases in both Gd and Pr sublattice sites with dilution, suggesting different mechanisms for Gd and Pr magnetic ordering.[15,16] In order to focus on the Pr magnetic ordering temperature without considering the possibility of phase separation in the $R_{1-x}Pr_xBa_2Cu_3O_{7-y}$ (R denotes rare earth) system,[4,8] we investigate the magnetic properties of metal (M) substituted $PrBa_2Cu_3O_{7-y}$. Substitution of transition metals introducing the structural disorder on either the Cu-O_2 planes or the Cu-O chains in $PrBa_2Cu_3O_{7-y}$ may offer an excellent opportunity to study the Pr-Pr magnetic coupling mechanism. Most recently, we have reported that the T_N of $PrBa_2Cu_3O_{7-y}$ remains essentially constant with Zn substitution, while it is reduced rapidly with increasing Ga substitution.[17] In this paper, we present the influence of the Fe, Co, and Ni substitution on the magnetic ordering and electrical resistivity of $PrBa_2Cu_3O_{7-y}$ and systematically discuss the correlation between them.

II. EXPERIMENTAL DETAILS

All polycrystalline samples were prepared by the standard solid-state-reaction method under identical conditions. High-purity Pr_6O_{11}, $BaCO_3$, CuO, and Fe_2O_3, CoO, or NiO powders were mixed and fired in air at 920 °C for 24 h. The resultant powders were pressed into pellets and heated in air at 920 °C for 24 h. This process was repeated at least four times with intermediate grinding. These pellets were then heated in flowing oxygen at 920 °C for 48 h followed by an additional anneal at 450 °C for 12 h. Finally the samples were slowly cooled in oxygen over several hours to room temperature. The structural analysis was carried out by the powder x-ray diffraction. Most samples showed single-phase oxygen-deficient perovskite structure. The lattice parameters were calculated from the diffraction peak positions by the method of least squares. The dc magnetization for each sample was measured using a superconducting quantum interference device (SQUID) magnetometer (quantum design) over the temperature range 4.2–40 K at a field of 1 kOe. The antiferromagnetic ordering temperature T_N was determined by the change of slope in the M-T or the relative minimum in the dM/dT-T curves. Electrical resistivity $\rho(T)$ measurements were performed on rectangular specimens cut from sintered pellets employing the standard four-probe method with silver paint contacts attached to electrical leads. Data were taken from 300 K down to 10 K.

III. RESULTS AND DISCUSSION

The lattice parameters a and b as a function of doping concentration x $(0 \leq x \leq 0.2)$ for $PrBa_2(Cu_{1-x}M_x)O_{7-y}$ with $M =$Fe, Co, and Ni are shown in Fig. 1. All systems studied exhibit an orthorhombic-tetragonal transition $(a = b)$ at about $x = 0.025$ for Fe and Co, and $x = 0.05$ for Ni. No marked change in slope of the c axes vs x (not shown) is observed. It is commonly agreed that the Ni and Zn substitute preferentially on the Cu-O_2 planes [Cu(2)], and Fe, Co, and Ga prefer the Cu-O chains [Cu(1)] in the $YBa_2(Cu_{1-x}M_x)_3O_{7-y}$ system. Such preferential substitution is accommodated by the different valence states of Fe, Co, Ni, Zn, and Ga. The Cu(2) site has a valence of 2+, well suited for the Ni^{2+} and Zn^{2+} ions. the Cu(1) site, may be in the 3+ state or in the mixed-valence state of 3+ and 2+, making it more favorable for the Fe^{3+}, Co^{3+}, and Ga^{3+} ions. This similarity in structural transition for Fe-, Co-, and Ga-doped $YBa_2Cu_3O_{7-y}$ and $PrBa_2Cu_3O_{7-y}$ suggests that Fe, Co, and Ga are also more favorable for trivalent ions and the Cu-O chains in the latter compound. The difference in structural transition between $YBa_2Cu_3O_{7-y}$ and $PrBa_2Cu_3O_{7-y}$ with Ni and Zn doping, where the formers remain orthorhombic throughout their range of existence,[18] whereas the latters undergo an orthorhombic-tetragonal transition at very low doping concentration ($x \sim 5$ at. %), may be due to the less orthorhombicity of $PrBa_2Cu_3O_{7-y}$ than $YBa_2Cu_3O_{7-y}$ or at the vicinity of phase instability for $PrBa_2Cu_3O_{7-y}$. The order of critical concentration x, where the orthorhombic-tetragonal transition occurs, is x(Fe, Co, and Ga doped $PrBa_2Cu_3O_{7-y}$)$\sim 0.025 <$ x(Fe-, Co-, and Ga-doped $YBa_2Cu_3O_{7-y}$) (Ref. 18) $\sim 0.05 \leq$ x(Ni and Zn doped $PrBa_2Cu_3O_{7-y}$)~ 0.05, supporting the above assumption. Tarascon *et al.*[18] reported that in $YBa_2Cu_3O_{7-y}$ the oxygen content increased with increasing Fe, Co, and Al, whereas it remained roughly constant and slightly decreases with increasing concentration of Ni and Zn, respectively. In addition, the oxygen content was observed to keep basically constant in the $Y_{1-x}Pr_xBa_2Cu_3O_{7-y}$ series.[8,19] Therefore, the oxygen content of the systems studied is thought to be close to 7.0 under our sample preparation conditions. Although $T_N = 14$ K of $PrBa_2Cu_3O_{6.46}$ (Ref. 20) and $T_N = 13$ K of $PrBa_2Cu_3O_6$ (Ref. 21) were reported, it is still reasonable to believe that the variation in magnetic ordering temperature of these metal-substituted $PrBa_2Cu_3O_{7-y}$ arises mainly from the intrinsic electronic structure of Fe, Co, Ni, Zn, and Ga.

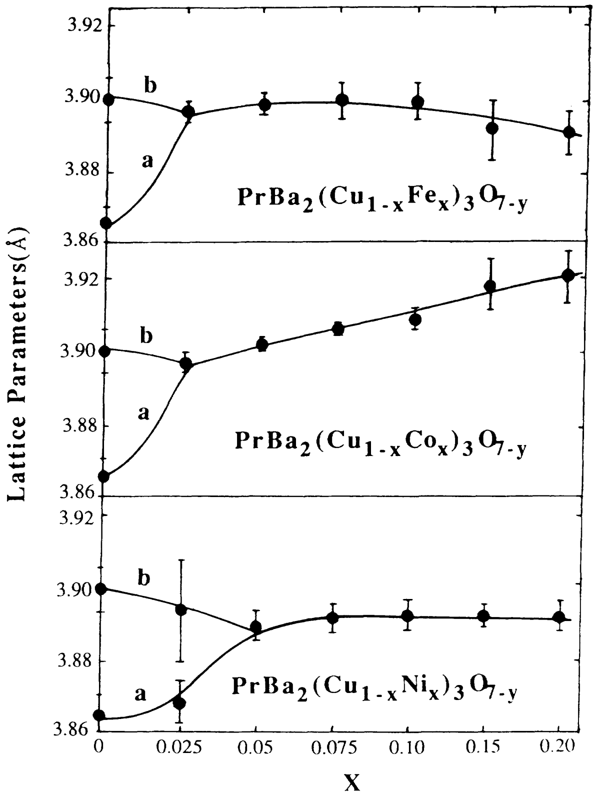

FIG. 1. Lattice parameters a and b as a function of x $(x \leq 0.2)$ for $PrBa_2(Cu_{1-x}M_x)_3O_{7-y}$ series with $M =$Fe, Co, and Ni. Lines are guides for the eye.

The magnetization M and the temperature derivative of magnetization dM/dT as a function of temperature for $PrBa_2(Cu_{1-x}M_x)_3O_{7-y}$ ($M =$Fe, Co, and Ni) with $0 \leq x \leq 0.2$ are shown in Figs. 2, 3, and 4, respectively. It is noted that the absolute values of M and dM/dT in these figures have been shifted for clarity. The $M(T)$ continues to increase as the temperature is lowered and a change of slope occurs at about 17.5 K (for $x = 0$), which is taken as an antiferromagnetic ordering temperature T_N. The dM/dT vs T curve also shows an anomaly at the same temperature. The continued increase of $M(T)$ for $T < T_N$ is quite unusual and may be related to Pr paramagnetism superimposed on the Cu-plane and/or chain ordering. A fluctuating behavior observed in dM/dT-T curves for Fe- and Ni-doped samples at high concentrations $(x > 0.10)$ may be due to impurity, which is an excess of the solubility limit. The T_N decreases and the magnetic anomaly is smeared monotonically with increasing doping concentration for all three systems. The doping-concentration dependence of T_N for $PrBa_2(Cu_{1-x}M_x)_3O_{7-y}$ ($M =$Fe, Co, Ni, Zn, and Ga) is shown in Fig. 5. The obtained depression rate dT_N/dx for these systems for $x \leq 0.1$ is about 0.65(Fe), 0.75(Co), 0.45(Ni), 0(Zn), and 1.2(Ga) K/at. %. The interpretation[17] given for the variation of T_N in Zn- and Ga-doped samples can be also applied to these Fe-, Co-, and Ni-doped systems quite successfully. That is, the Cu-O chains play an important role in $PrBa_2Cu_3O_{7-y}$ magnetic ordering. The Zn-doped system showed no significant change in T_N while the Ga-doped system had the largest depression in T_N indicating that the Zn and Ga occupied the Cu(2) and Cu(1) sites, respectively. It also suggests that the Zn is divalent and the Ga is trivalent. The magnitude of the depression rate in T_N by the Fe-, Co-, and Ni-doped systems is ascribed mostly to the different percentage of these metals occupying the Cu(1) sites. To coincide with the results in Fig. 5, the substitution of Fe, Co, and Ni most likely occurs on both Cu sites. For instance, the dT_N/dx of Ni-doped samples is nonlinear, suggesting that at higher Ni doping the Ni tends to occupy Cu(2) sites, thus slowing the depression. It is noted that the dT_N/dx does not appear to be affected significantly by the orthorhombic-tetragonal transition which occurs at $x=0.025$ for Fe and Co, and at $x=0.5$ for Ni. Lopex-Morales *et al.*[20] also found the onset of an orthorhombic-tetragonal transition as the oxygen content of $PrBa_2Cu_3O_{7-y}$ drops below 6.6, while its magnetic transition temperature is in the region of 14 to 17 K. These may indicate that the T_N of $PrBa_2Cu_3O_{7-y}$ is more sensitive to local Cu(1) site disorder than to long-range lattice structure. From these results, we should be able to simply conclude that the more Cu(1) site disorder is introduced, the more effective suppressing of T_N is observed. As we just mentioned above,[18] substitutions for the different Cu sites are generally accompanied by a change of oxygen content. Therefore the small variation of oxygen content for these systems caused by different percentage of metals in the Cu(1) site somehow would affect the depression rate. Though the influence by the different magnetic moments of doping metals may be small and is hard to separate from the others, a minor effect on T_N will be expected.

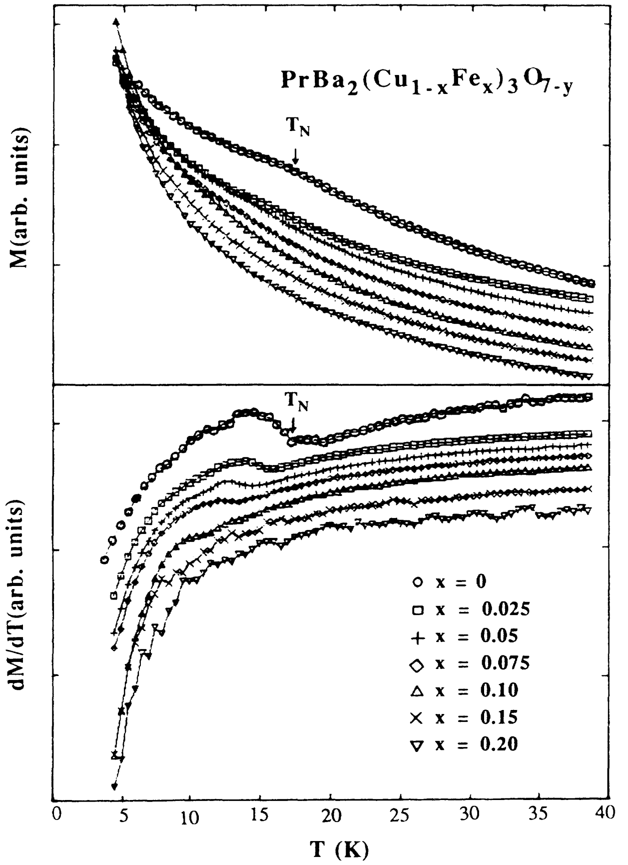

FIG. 2. Low-temperature magnetization $M(T)$ (taken at $H=1$ kOe) and the temperature derivative of magnetization dM/dT vs temperature for $PrBa_2(Cu_{1-x}Fe_x)_3O_{7-y}$ with $x=0$, 0.025, 0.05, 0.075, 0.10, 0.15, and 0.20. For clarity, the M and dM/dT have been shifted by an appropriate value. T_N determined from the change of slope in the $M(T)$ or the relative minimum in the dM/dT curves is indicated by an arrow.

In order to elucidate the unusual magnetic properties of $PrBa_2Cu_3O_{7-y}$, we summarize the substitution effect on the superconducting transition and magnetic ordering temperature for some selected compounds in Table I. There are several comparisons related to this work, some of which might have been concluded in quoted references, worth being emphasized from the table. (1) The T_c is suppressed with increasing metal doping concentration and is more sensitive to local structural disorder than to magnetic interaction.[18] For example, the nonmagnetic

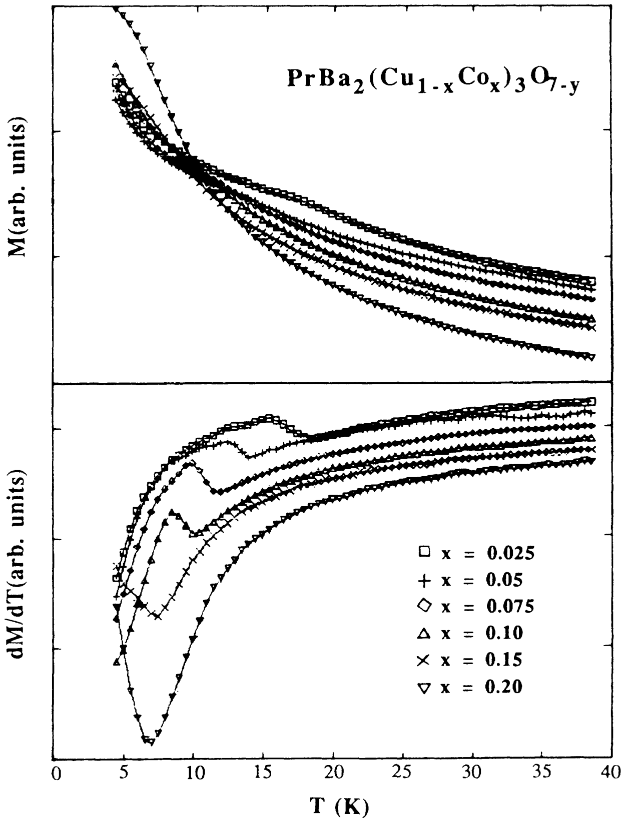

FIG. 3. Low temperature magnetization $M(T)$ and the temperature derivative of magnetization dM/dT vs temperature for $PrBa_2(Cu_{1-x}Co_x)_3O_{7-y}$ with $x=0.025$, 0.05, 0.075, 0.10, 0.15, and 0.20.

TABLE I. Substitution effect on the superconducting transition and magnetic ordering temperature for some selected compounds at the doping concentration $x=0.1$.

Compound	$T_c(0)$ (K)	$T_N(0)$ (K)	dT_c/dx (K/at. %)	dT_N/dx (K/at. %)	Reference
$YBa_2(Cu_{1-x}Fe_x)_3O_{7-y}$	94		−5.6		28
$YBa_2(Cu_{1-x}Co_x)_3O_{7-y}$	94		−7.3		28
$YBa_2(Cu_{1-x}Ni_x)_3O_{7-y}$	94		−2.8		28
$YBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$	94		−13		29
$YBa_2(Cu_{1-x}Ga_x)_3O_{7-y}$	94		−1.1		29
$GdBa_2(Cu_{1-x}Fe_x)_3O_{7-y}$		2.25		~0	23
$GdBa_2(Cu_{1-x}Co_x)_3O_{7-y}$		2.25		~0	23
$GdBa_2(Cu_{1-x}Ni_x)_3O_{7-y}$		2.25		−0.03	22
$GdBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$		2.25		−0.02	22
$PrBa_2(Cu_{1-x}Fe_x)_3O_{7-y}$		17.5		−0.65	This work
$PrBa_2(Cu_{1-x}Co_x)_3O_{7-y}$		17.5		−0.75	This work
$PrBa_2(Cu_{1-x}Ni_x)_3O_{7-y}$		17.5		−0.45	This work
$PrBa_2(Cu_{1-x}Zn_x)_3O_{7-y}$		17.5		~0	17
$PrBa_2(Cu_{1-x}Ga_x)_3O_{7-y}$		17.5		−1.2	17
$Pr_{1-x}Y_xBa_2Cu_3O_{7-y}$		17		−0.25	7
$Pr_{1-x}Gd_xBa_2Cu_3O_{7-y}$		17		−0.25	16
$Y_{1-x}Pr_xBa_2Cu_3O_{7-y}$	90		−1.15		7
$Gd_{1-x}Pr_xBa_2Cu_3O_{7-y}$	92	2.25	−1.8	−0.02	16

Zn ion has a larger effect on T_c than magnetic Fe, Co, and Ni ions. (2) The effect of metal substitution on T_N strongly depends on which Cu sites the metal occupies. The T_N of $GdBa_2Cu_3O_{7-y}$ is more affected when the doping occurs in the Cu-O_2 planes[22] (such as Ni and Zn) than in the Cu-O chains (such as Fe and Co).[23] On the other hand, results from this work on $PrBa_2Cu_3O_{7-y}$ exhibit an almost opposite behavior. In addition, the T_N of $GdBa_2Cu_3O_{7-y}$ was not significantly dependent on the oxygen content,[24] but a small effect for $PrBa_2Cu_3O_{7-y}$.[20,21] More importantly, the $GdBa_2Cu_3O_7$ is a metal (where RKKY could apply), whereas the

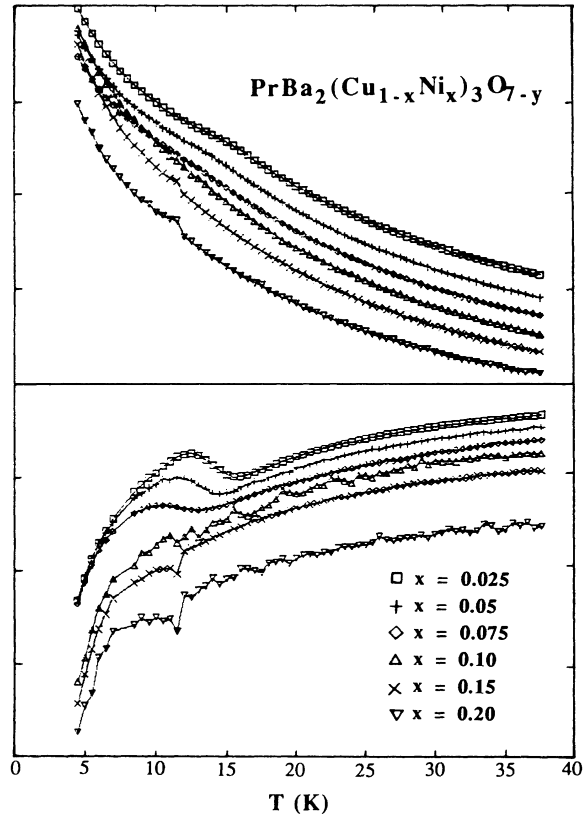

FIG. 4. Low-temperature magnetization $M(T)$ and the temperature derivative of magnetization dM/dT vs temperature for $PrBa_2(Cu_{1-x}Ni_x)_3O_{7-y}$ with $x=0.025$, 0.05, 0.075, 0.10, 0.15, and 0.20.

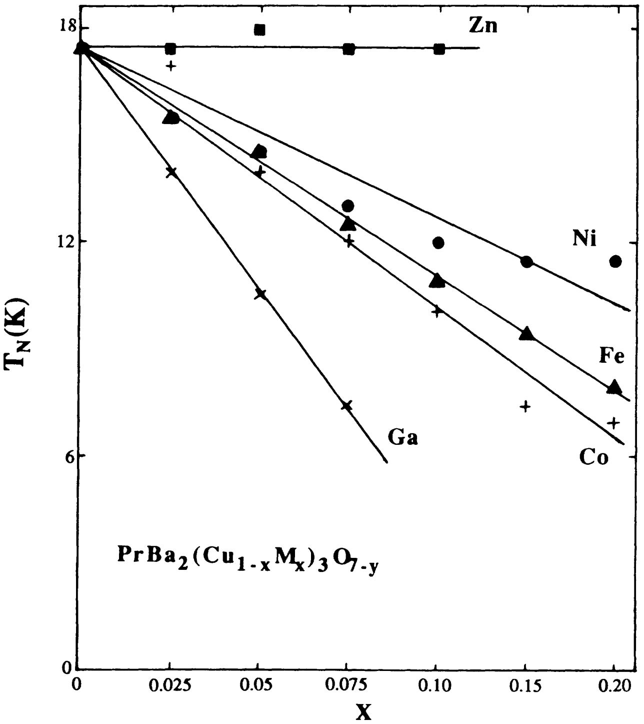

FIG. 5. Composition dependence of the antiferromagnetic ordering temperature T_N for $PrBa_2(Cu_{1-x}M_x)_3O_{7-y}$ with $M=$Fe, Co, Ni, Zn, and Ga. Lines are guides for the eye.

$PrBa_2Cu_3O_{7-y}$ is an insulator (which superexchange may dominant in Pr-Pr coupling). These observations suggest the different magnetic coupling mechanisms in Gd and Pr magnetic order. In fact, it is difficult to explain that the Cu-O_2 planes play the equally important role both in high-T_c superconductivity and low-temperature magnetic order of $GdBa_2Cu_3O_{7-y}$ simultaneously, while the removal of oxygen only suppresses superconductivity. This might be the reason why the different coupling mechanisms in Gd magnetic order are being debated.[16,22–25] (3) Because the distance of the Pr-CuO_2 plane is smaller than that of Pr-CuO chain in the unit cell, the magnetic coupling of Pr-Pr via the Cu-O chain would be more likely a three-dimensional antiferromagnetic characteristic. This consideration is consistent with the large T^3 term in low-temperature specific-heat measurements.[12,26] (4) The almost equal depression rate in T_N ($\sim$0.25 K/at. %) of $PrBa_2Cu_3O_{7-y}$ by the doping of Y and Gd indicates that the Pr and Gd ions act independently. Thus the direct intersite $4f$-$4f$ magnetic interaction appears to be weak. Thereby the question of why the T_N of $PrBa_2Cu_3O_{7-y}$ is as high as 17 K is a challenge for theorists. (5) The Pr is more effective in depressing T_c in $GdBa_2Cu_3O_{7-y}$ ($\sim$1.8 K/at. %) than in $YBa_2Cu_3O_{7-y}$ ($\sim$1.15 K/at. %). This result can be explained by assuming the strong hybridization between extended Pr $4f$-electrons and conduction bands in the Cu-O_2 planes. This unique electronic characteristic of Pr should be taken into account for the quenching of superconductivity and the anomalous magnetic properties of $PrBa_2Cu_3O_{7-y}$.

Although the mechanism for the depression of superconductivity in $Y_{1-x}Pr_xBa_2Cu_3O_{7-y}$ is being debated in the current literatures[1–13] with the quenching of mobile holes in the Cu-O_2 planes via either the hole-filling[3,7] or the localization of states due to disorder,[5,6] all the remaining charges of $PrBa_2Cu_3O_{7-y}$ should reside in the Cu-O chains. Due to the frozenness of plane conductivity in $PrBa_2Cu_3O_{7-y}$, it provides an excellent chance to isolate and study the chain transport specifically. So, we measure the electrical resistivity for these systems to study more about the correlation between the magnetic ordering and Cu-O chains. The temperature dependence of resistivity of $PrBa_2(Cu_{1-x}M_x)_3O_{7-y}$ as a function of doping metal (Fe, Co, and Ni) concentration is shown in Fig. 6. Clearly, the semiconducting resistivity increases rapidly with increasing Fe and Co concentration, while it changes slightly with Ni-doped samples at $x \geq 0.025$. This observation confirms the suggestion that the residual electrical transport of $PrBa_2Cu_3O_{7-y}$ comes mostly from the Cu-O chains. Thus substitutions on the Cu(2) sites (such as Zn or Ni) do not affect the resistivity as much as those on the Cu(1) sites (such as Fe, Co, and Ga). In fact, the resistivity changes for Ni are nonmonotonic, whereas

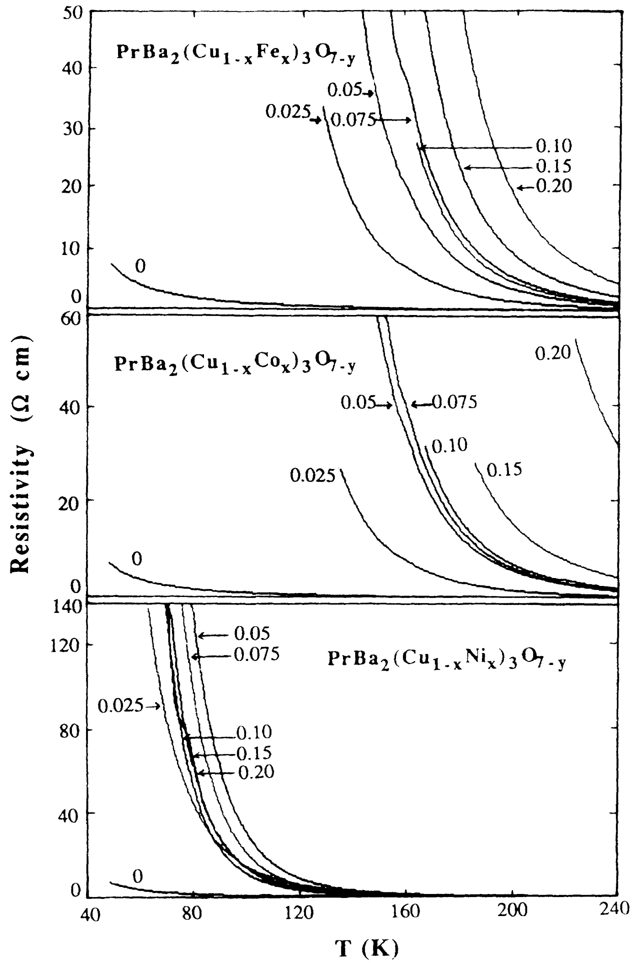

FIG. 6. Temperature dependence of the resistivity of $PrBa_2(Cu_{1-x}M_x)_3O_{7-y}$ as a function of doping metal (Fe, Co, and Ni) concentration for x = 0, 0.025, 0.05, 0.075, 0.10, 0.15, and 0.20.

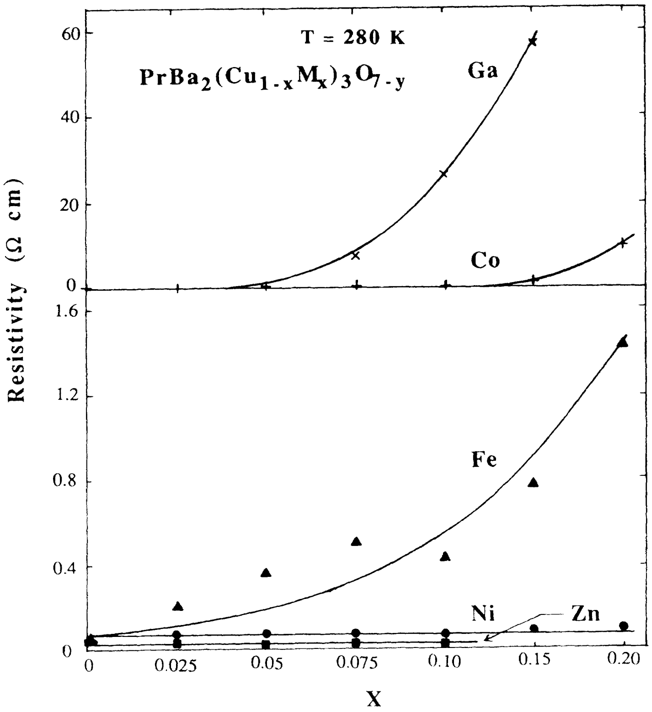

FIG. 7. Composition dependence of room-temperature resistivity $\rho_{280\,K}$ for $PrBa_2(Cu_{1-x}M_x)_3O_{7-y}$ with M = Fe, Co, Ni, Zn, and Ga. Lines are guides for the eye. It is noted that the scales of resistivity are different.

they are monotonic for the others. This should be related to the occupancy of Ni both in Cu(2) and Cu(1) sites which we have mentioned above. Composition dependence of room-temperature resistivity $\rho_{280\,K}$ for $PrBa_2(Cu_{1-x}M_x)_3O_{7-y}$ with $M =$ Fe, Co, Ni, Zn, and Ga are displayed in Fig. 7. The striking feature is that the increasing rate $d\rho/dx$ at $T=280$ K is $Zn < Ni < Fe < Co < Ga$ at all studied doping concentrations. This relation is completely corresponding to the depression rate in T_N found in Fig. 5. Consequently, we conclude that the more Cu(1) site disorder is introduced, the more increase in resistivity and the more effective in suppressing T_N are observed in $PrBa_2Cu_3O_{7-y}$.

It should be remembered that the muon-spin-relaxation (μSR) results on $PrBa_2Cu_3O_{7-y}$ did not rule out the possibility of magnetic ordering ($T_N=17$ K) of Cu moments within the Cu-O chains.[10] No magnetic ordering of Pr moments was observed[27] above 1.5 K in the parent compound Pr_2CuO_{4-y}, where there is no Cu-O chain and only Cu-O_2 planes, suggesting some speculations on this point of view. If it is Cu(1) that orders, our metal-substituted results can still explain that. That is, the depression rate in T_N of $PrBa_2Cu_3O_{7-y}$ depends on the Cu(1)-sites occupancy of doping metal. The more Cu(1)-site disorder is introduced, the more effective in suppressing T_N is observed. In such a case, how can the Cu(1) moments ordering explain the existing anomalous magnetic properties of $PrBa_2Cu_3O_{7-y}$? If it is possible and needed, a study of competition between superconductivity and antiferromagnetism in Cu(1) and Cu(2) sites of $Y_{1-x}Pr_xBa_2(Cu_{1-y}M_y)_3O_{7-z}$ will be very attractive.

In summary, effects of Fe, Co, and Ni substitution on magnetic ordering and electrical resistivity of $PrBa_2Cu_3O_{7-y}$ have been studied and compared to those of related compounds. An almost opposite behavior in depression of T_N by the metal (Fe, Co, Ni, Zn, and Ga) substitution observed in $PrBa_2Cu_3O_{7-y}$ and $GdBa_2Cu_3O_{7-y}$ indicates possibly different magnetic coupling mechanisms in these two compounds. The more Cu(1)-site disorder is introduced, the more effective in suppressing T_N and the more insulating behavior are observed in $PrBa_2Cu_3O_{7-y}$. Combining the results of structural analysis, depression rate in T_N, and electrical resistivity, we suggest that the Cu-O chain transport or the oxygen-vacancy order would play a key role in magnetic order of $PrBa_2Cu_3O_{7-y}$.

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