PHYSICAL REVIEW B VOLUME 59, NUMBER 17 1 MAY 1999-I

Pressure effects on the superconductivity and magnetic order of $Y_{1-x}Pr_xBa_2Cu_4O_8$

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(Received 11 May 1998; revised manuscript received 11 January 1999)

Pressure effects on the superconductivity and magnetic order of polycrystalline $Y_{1-x}Pr_xBa_2Cu_4O_8$ ($0 \leqslant x \leqslant 1$) were investigated. It was found that T_c decreases monotonically with Pr concentration x from $T_c \sim 80$ K for $x=0$ to $T_c \sim 6$ K for $x=0.7$. The critical Pr concentration x_{cr} required to suppress the T_c to zero is estimated to be around 0.72, which is larger than that (~ 0.55) in $Y_{1-x}Pr_xBa_2Cu_3O_7$. The normal-state resistivity remains in the same order of magnitude through whole series in contrast to a metal-insulator transition observed in $Y_{1-x}Pr_xBa_2Cu_3O_7$. A change of sign in the value of $(dT_c/dP)_{P=0}$ in $Y_{1-x}Pr_xBa_2Cu_4O_8$ with Pr doping is observed which is similar to that of $Y_{1-x}Pr_xBa_2Cu_3O_7$. The T_N in $PrBa_2Cu_4O_8$ decreases with increasing external pressure as in $PrBa_2Cu_3O_7$. The results for T_c and T_N variations with Pr doping and pressure effect in $Y_{1-x}Pr_xBa_2Cu_4O_8$ cannot be fully explained by the recent hybridization theory. [S0163-1829(99)03217-8]

It is well known[1,2] that in $YBa_2Cu_3O_{7-\delta}$ (Y123), the substitution of Pr for Y destroys superconductivity while other rare-earth elements, even if they are magnetic do not affect superconductivity.[3–5] Despite the large number of experimental and theoretical works that have been done, the depression of superconductivity in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ [(Y,Pr)123] is still not completely understood. Moreover, the pressure dependence of superconducting transition temperature T_c for Pr-doped Y123 behaves anomalously.[6] However, very recently it has been reported that Pr123 single crystals grown by the traveling-solvent floating-zone method show superconductivity and T_c enhancement under pressure.[7,8] This absence or occurrence of superconductivity in Pr123 has made the issue more interesting and reopened the field for discussion. As a consequence, the substitution of Pr for Y in the $YBa_2Cu_4O_8$ (Y124) has also received much attention. Several groups[9,10] have already reported on the $Y_{1-x}Pr_xBa_2Cu_4O_8$ [(Y,Pr)124], only with $x \leqslant 0.8$, because of the unsuccessful synthesis of $PrBa_2Cu_4O_8$ (Pr124) in previous years. Very recently, we have successfully synthesized Pr124 at ambient oxygen pressure.[11,12] In the present report, we have systematically studied the pressure effect on T_c and T_N of the $Y_{1-x}Pr_xBa_2Cu_4O_8$ (with $0 \leqslant x \leqslant 1$) in order to better understand the T_c suppression and magnetic coupling mechanisms of high T_N in these Pr-doped systems.

The sample preparation for $Y_{1-x}Pr_xBa_2Cu_4O_8$ ($0 \leqslant x \leqslant 1$) was described else where.[11,12] It is noted that the annealing temperature is 800 °C for $0 \leqslant x \leqslant 0.4$ and 870 °C for $0.6 \leqslant x \leqslant 1$. The structural analysis was carried out by powder x-ray diffraction using Cu $K\alpha$ radiation. Low-angle ($2\theta = 5° - 10°$) x-ray diffraction and thermogravimetric analysis showed that all samples are basically single $YBa_2Cu_4O_8$ (124) phase without any indication of $YBa_2Cu_3O_7$ (123) impurity. But for samples with $x \geqslant 0.8$, very few impurity peaks from $BaCuO_2$, estimated to be less than 5%, were found. There is no correlation between the Pr content and the amount of impurity phase, indicating that the depression of superconductivity in the studied system does not result from the impurity phase. The a and b axes and unit-cell volume V are found to increase with x as has been found in (Y,Pr)123 (Ref. 5). Similar to (Y,Pr)123 (Ref. 5), the c axis in (Y,Pr)124 increases very slightly with x. The pressure dependences of T_c and T_N were determined for a bulk sample by a low-frequency (16.2 Hz) ac magnetic-susceptibility technique using a piston-cylinder type Be-Cu hydrostatic pressure clamp. 3M fluorinert liquid was used as the pressure transmitting fluid. Pressure was applied at room temperature and determined at low temperature by means of a superconducting lead manometer.

In Fig. 1 the resistivity is plotted as a function of temperature for different Pr concentrations in (Y,Pr)124. The samples with x up to 0.7 display metallic behavior in the

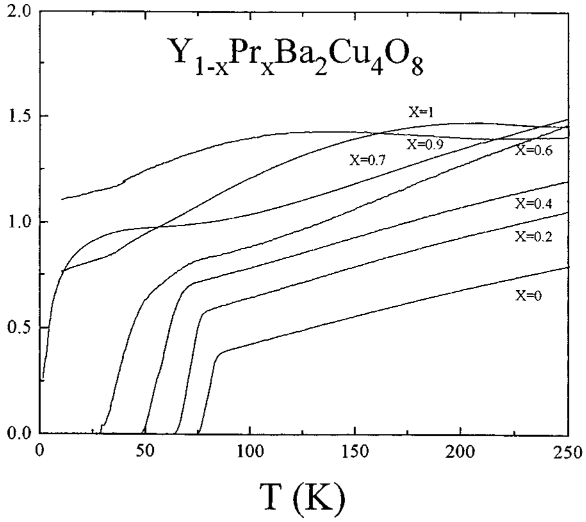

FIG. 1. Temperature variation of resistivity for $Y_{1-x}Pr_xBa_2Cu_4O_8$ with $x=0$, 0.2, 0.4, 0.6, 0.7, 0.9, and 1.0.

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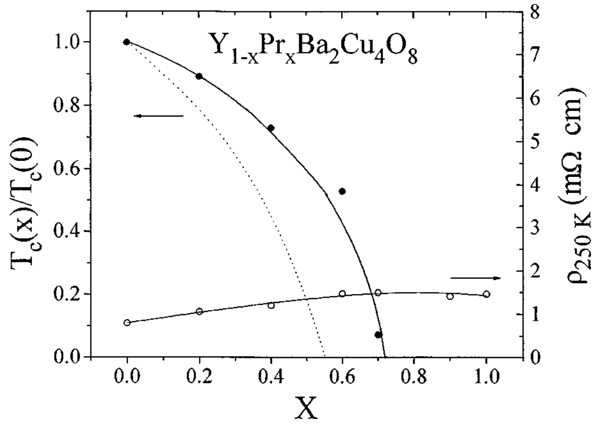

FIG. 2. The variations of $T_c(x)/T_c(0)$ and room-temperature resistivity ($\rho_{250\,K}$) with Pr content (x) for $Y_{1-x}Pr_xBa_2Cu_4O_8$. $T_c(0)$ is the zero-resistance temperature for $x=0$. The solid line drawn through the data is a guide to the eye. The dashed line is reproduced from Ref. 5 for $Y_{1-x}Pr_xBa_2Cu_3O_7$.

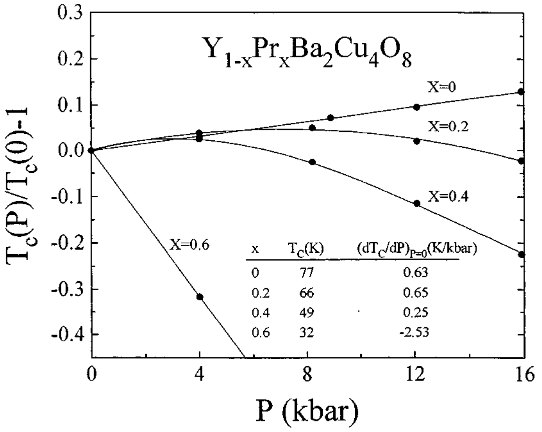

FIG. 3. $[T_c(P)/T_c(0)-1]$ as a function of hydrostatic pressure (P) between 0 and 16 kbar in the $Y_{1-x}Pr_xBa_2Cu_4O_8$ system with $x=0$, 0.2, 0.4, and 0.6. The values of T_c at ambient pressure $T_c(0)$ and the initial rate of change of T_c with pressure, $(dT_c/dP)_{P=0}$, for each value of x are listed in the figure. The lines drawn through the data are guides to the eye.

normal-state resistivity. For $x=0.9$ and 1.0 samples the resistivity takes a maximum, respectively, around 125 and 200 K, below which both the samples exhibit metallic conduction. This is in contrast to (Y,Pr)123 with $x\geqslant 0.5$, where the normal-state behavior is semiconducting.[5] The variation of $T_c(x)/T_c(0)$ as a function of x for (Y,Pr)124 is shown in Fig. 2, where, $T_c(0)$ is the T_c for $x=0$ sample and $T_c(x)$ is the T_c of the sample with corresponding x. It is found that the value of x_{cr} (critical concentration of Pr, where T_c disappears) for (Y,Pr)124 is estimated to be ~0.72 which is larger than that ($x_{cr}\sim 0.55$) of (Y,Pr)123. According to the hybridization theory proposed in Refs. 13 and 14, the larger x_{cr} in (Y,Pr)124 suggests that the hybridization has a weaker hole-depletion power than in (Y,Pr)123. However, both (Y,Pr)124 and (Y,Pr)123 contain same CuO_2-(Y,Pr)-CuO_2 layers, which play an important role in the hybridization theory. Therefore, it is difficult to explain this T_c depression, i.e., larger x_{cr} value in (Y,Pr)124 than that in (Y,Pr)123 by a hole-depletion mechanism, especially because Y124 is a underdoped cuprate. It is also noted that the x_{cr} value in $Y_{1-x}Pr_xSr_2Cu_{2.7}Mo_{0.3}O_7$ is ~0.85 (Ref. 15) and this system has the tetragonal symmetry instead of the orthorhombic structure of Y123. The different parameters for (Y,Pr)123, (Y,Pr)124, and $Y_{1-x}Pr_xSr_2Cu_{2.7}Mo_{0.3}O_7$ are listed in Table I for comparison. The normal-state resistivity (ρ) at 250 K as a function of x for (Y,Pr)124, shown in Fig. 2, increases slightly with increase of Pr content up to $x=0.7$, and is nearly constant for $x>0.7$. But in the (Y,Pr)123 system, resistivity increases rapidly for $x>0.55$ with increasing Pr content.[5] This is due to the double Cu-O chains in (Y,Pr)124 instead of a single Cu-O chain in (Y,Pr)123. It is also well established that both single and double Cu-O chains are metallic in Y123 and Y124 (Refs. 16–18). NMR and NQR measurements have shown that spin susceptibilities on the planes and chains are markedly different,[19–21] suggesting that the two conducting channels (CuO_2 sheet and CuO chain) may act independently of one another. Therefore, it is plausible that both the higher x_{cr} and the absence of the metal-insulator transition in (Y,Pr)124 are due to the double CuO chains.

The $T_c(P)$ data obtained from the temperature dependence of ac susceptibility under different hydrostatic pressures for (Y,Pr)124 samples are displayed in Fig. 3, where the change in T_c with P is depicted as $[T_c(P)/T_c(0)\text{-}1]$ for clarity, and $T_c(0)$ is the T_c at ambient pressure. These data reveal the following features: The Y124 compound exhibits a linear increase of T_c with P to ~16 kbar at the rate $dT_c/dP=0.63$ K/kbar. This is in reasonable agreement with a previously reported value of 0.66 K/kbar (Ref. 22). The large positive dT_c/dP observed for Y124 has been interpreted to be caused by carrier transfer from the double CuO chains to the CuO_2 sheets in this underdoped material.[23,24] This interference is supported by high pressure neutron-diffraction refinement.[25,26] The sample with $x=0.2$ shows positive initial slope (0.65 K/kbar) and displays a distinct maximum at ~6 kbar followed by a decrease. The sample with $x=0.4$ shows a maximum at ~2.5 kbar, suggesting the

TABLE I. Superconducting transition temperature T_c, critical Pr concentration x_{cr} where T_c disappears, antiferromagnetic transition temperature T_N, Pr concentration x_{cp} where $(dT_c/dP)_{p=0}$ changes sign from positive to negative and dT_N/dP for $Y_{1-x}Pr_xBa_2Cu_3O_7$, $Y_{1-x}Pr_xBa_2Cu_4O_8$ and $Y_{1-x}Pr_xSr_2Cu_{2.7}Mo_{0.3}O_7$ compounds.

Compound	T_c $(x=0)$	x_{cr}	T_N $(x=1)$	x_{cp}	dT_N/dP	Ref.
$Y_{1-x}Pr_xBa_2Cu_3O_7$	93 K	0.55	17 K	~0.35	−1 K/kbar	7, 27, 28
$Y_{1-x}Pr_xBa_2Cu_4O_8$	80 K	0.72	17 K	>0.4	−1.1 K/kbar	12, this work
$Y_{1-x}Pr_xSr_2Cu_{2.7}Mo_{0.3}O_7$	33 K	0.85	<0.5 K			15

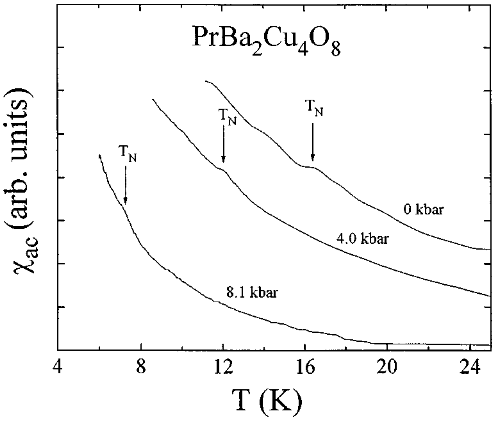

FIG. 4. Temperature variation of χ_{ac} of $PrBa_2Cu_4O_8$ at different hydrostatic pressures. The arrows indicate the antiferromagnetic ordering temperature (T_N).

occurrence of a pressure-induced change of electronic property in the vicinity of 2.5 kbar. The samples with $x>0.4$ show a rapid monotonic decrease of T_c with P. These data imply that the increase of Pr concentration x has the effect of shifting the maximum of $T_c(P)$ curve from higher to lower pressures and eventually to zero pressure above $x=0.4$. At this point, $(dT_c/dP)_{P=0}$ changes sign from positive to negative. Neumeier *et al.*[27] explained $T_c(P)$ data for (Y,Pr)123 considering the hybridization of the Pr $4f$ electrons and plane O $2p$ states. It is noted that $(dT_c/dP)_{P=0}$ of (Y,Pr)123 changes sign in between $x=0.3$ and 0.4, which is lower than that of (Y,Pr)124 (in this case sign changes above $x=0.4$). Therefore, if the hybridization scenario is suitable to explain the $T_c(P)$ both in (Y,Pr)124 and (Y,Pr)123, it seems that the pressure-induced enhancement of hybridization is weaker in (Y,Pr)124 than in (Y,Pr)123. On the other hand, for $x=0.2$ the $T_c(P)$ in (Y,Pr)123 shows maximum at pressure >16 kbar which is higher than that (~6 kbar) in (Y,Pr)124. From this point of view, the hybridization mechanism may not fully explain the $T_c(x,P)$ behavior in the (Y,Pr)124 system.

The anomaly observed in the $\chi_{ac}(T)$ curve (Fig. 4) is due to the antiferromagnetic ordering of Pr sublattices. It is found that in Pr124 the antiferromagnetic ordering temperature (T_N) decreases with the increase of pressure as found in Pr123 (Ref. 28). In both the cases the value of T_N (~17 K) and the pressure depression rate $dT_N/dP(\sim -1$ K/kbar) are nearly identical and anomalously high. Therefore, magnetic coupling of Pr-containing cuprates may be related to CuO_2 planes rather than CuO chains. In fact, the issue of the correlation between T_c and T_N in Pr-based cuprates has long been interesting and controversial.[29] In the case of single CuO chain containing Pr-based cuprates, it has been discussed earlier[30] that a strong magnetic correlation, which is caused or mediated by an enhanced hybridization of Pr $4f$ electrons with holes in the CuO_2 planes, is the primary origin for the suppression of T_c. But in the present investigation, it is found that in Pr-rich (Y,Pr)124 system, which contains double CuO chains, both T_c and T_N are suppressed with the increase of pressure. Furthermore, the T_N for both Pr123 and Pr124 is around 17 K, while, no magnetic ordering transition is found in $PrSr_2Cu_{2.7}Mo_{0.3}O_7$ (see Table I). Therefore, the correlation between T_c suppression and high T_N becomes unclear in the present state.

In summary, $T_c(x,P)$ in (Y,Pr)124 and $T_N(P)$ in Pr124 have been studied and compared to those in (Y,Pr)123 and Pr123, respectively. The T_c decreases at a slower rate with increasing Pr concentration x in (Y,Pr)124 than that in (Y,Pr)123. There is no evidence for a metal-insulator transition in (Y,Pr)124 in contrast to a metal-insulator transition occurring at $x\sim 0.55$ in (Y,Pr)123. This may be due to the double CuO chains of (Y,Pr)124 instead of a single CuO chain in (Y,Pr)123. Therefore, CuO chains should play a role in both the normal-state conductivity and superconductivity. At least three points are available from the data to argue that the hybridization mechanism cannot fully explain the $T_c(x,P)$ and $T_N(P)$ behavior of (Y,Pr)124:

(1) From the x_{cr} values it seems that the hybridization is weaker in (Y,Pr)124 ($x_{cr}\sim 0.72$) than in (Y,Pr)123 (x_{cr} ~ 0.55). This can explain the sign change of $(dT_c/dT)_{P=0}$ at $x>0.4$ for (Y,Pr)124 and in between $x=0.3$ and 0.4 for (Y,Pr)123 but cannot explain T_c maxima for $x=0.2$ samples which occur at $P=6$ kbar in (Y,Pr)124 and at $P>16$ kbar in (Y,Pr)123.

(2) If the hybridization is weaker in (Y,Pr)124 than in (Y,Pr)123, then nearly identical values of T_N and dT_N/dP in Pr124 and Pr123 cannot be explained.

(3) If the hybridization occurs in CuO_2 planes, it may reduce T_c and enhance T_N in Pr-rich samples. In that case it is difficult to explain the data in the present investigation where external pressure suppresses both T_c and T_N in Pr-rich samples.

This work was supported by National Science Council of Republic of China under Contracts No. NSC88-2112-M-110-007 and NSC88-2112-M-009-025.

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