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Magnetic order and superconductivity in $Y_{1-x}Pr_xBa_2Cu_3O_7$

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The system $Y_{1-x}Pr_xBa_2Cu_{3-y}Fe_yO_z$, with $x=0$ to 1, $y=0$ and 0.3, and $z=6$ and 7, has been studied by ac and dc magnetometry to determine the superconducting phase transition temperature, T_c, by ^{57}Fe Mössbauer spectroscopy to determine the antiferromagnetic phase transition temperature of the Cu(2) site, T_N, and by specific-heat measurements to determine γ and the antiferromagnetic ordering temperature of the Pr site, T_n. T_c $(x,z=7)$, T_N $(x,z=6,7)$, and T_n $(x,z=7)$ were obtained for x values of 0.0, 0.2, 0.4, 0.6, 0.8, and 1.0. For $z=6$, T_N changes very little with x [$T_N(0,6)=415$ K and $T_N(1,6)=350$ K], but for $z=7$, T_N changes sharply with x [$T_N(1,7)=325$ K, $T_N(0.6,7)=230$ K, and $T_N(0.4,7)\leq 35$ K]. There is no conclusive evidence for overlap of superconductivity and antiferromagnetism. T_n also decreases strongly with decreasing x, $T_n(1,7)=17$ K, while $T_n(0.4,7)\leq 2.5$ K. The specific-heat measurements show large linear temperature terms. The slope γ increases strongly with x and with the introduction of the Fe probe.

I. INTRODUCTION

It is well established that in the orthorhombic high-T_c superconductors $YBa_2Cu_3O_z$ [for $z=7$ and $T_c=92$ K (Ref. 1)], T_c is dramatically affected by oxygen content and distribution. When samples are heat treated at elevated temperatures and/or in a reducing atmosphere, both the oxygen content and T_c decrease. For $6.0<z<6.5$ the samples are tetragonal and semiconducting, and the Cu(2) sites are antiferromagnetically ordered. T_N increases sharply with decreasing z from $T_N=0$ K for $z=6.5$ to $T_N=415$ K for $z=6.0$.[2] The main question is whether this superconducting-magnetic phase diagram is unique and depends only on the oxygen concentration, or whether substitution of other elements into this system would lead to similar phase diagrams. Some of the very first experiments on the $Y_{1-x}R_xBa_2Cu_3O_7$ (R denotes rare earth) system showed that T_c did not depend significantly on the concentration x of trivalent rare-earth ions (e.g., Gd, Dy, Nd, etc.) in the material, indicating that the rare earth and the copper Cu(2)-O layers are electronically decoupled for all practical purposes.[3] An important exception to this behavior is Pr, which forms the same orthorhombic structure, but T_c is reduced sharply with increasing x,[4,5] and for $x>0.55$ the materials are not superconducting. Moreover, two magnetically ordered sublattices are reported for $PrBa_2Cu_3O_7$. Using the Mössbauer technique on iron-doped samples, we showed in a previous paper[6] that the Cu(2) sites are magnetically ordered, with $T_N=325$ K, even though the material is oxygen rich. This shows that the number of oxygen ions alone cannot account for the occurrence or disappearance of the magnetic interactions in the Cu(2) sites. The second antiferromagnetic ordering, that of the Pr sublattice at 17 K found by specific-heat susceptibility and neutron diffraction measurements, is almost 2 orders of magnitude higher than expected,[7] in comparison with the other rare-earth systems.

It has been reported by several groups that partial substitution of Cu in the orthorhombic superconductor $YBa_2Cu_2O_7$ by Fe progressively decreases T_c of the system; for iron concentration exceeding 13 at. % the materials are not superconducting.[8,9] Mössbauer and neutron diffraction measurements definitely show that the Fe ions substitute preferentially for the Cu in the Cu(1) sites and only about 10–20 % of Fe substitute for the Cu in the Cu(2) sites.[6] Moreover, the magnetic order which is enhanced with decreasing oxygen content in the samples is not affected by the presence of Fe,[6] and the Fe is thus a reliable probe of the magnetic behavior of the Cu(2) ions.

Here we present detailed studies of $Y_{1-x}Pr_xBa_2Cu_3O_z$ and $Y_{1-x}Pr_xBa_2Cu_{2.7}Fe_{0.3}O_z$ with $x=0$, 0.2, 0.4, 0.6, 0.8, and 1, and $z=6$ and 7, by several experimental techniques, and obtain detailed phase diagrams which show regions of superconductivity and regions of magnetic order on both Cu(2) and Pr sites. The systems exhibit either superconductivity for $x<0.55$ and $x<0.4$ for pure and Fe-doped materials, respectively, or antiferromagnetism in both Cu(2) and Pr sites for $x>0.55$. In the 10 at. % Fe-doped samples, T_N of the Cu(2) sites as determined by Mössbauer spectroscopy for $z=7.1$ is 325 K for $x=1$, 300 and 230 K for $x=0.8$ and 0.6, and drops to below 35 K for $x=0.4$. The magnetic transition temperature of the Pr site also decreases with decreasing x, $T_n=17$ K for $x=1$, but 12 K and 8 K for $x=0.8$ and 0.6, respectively. It appears that iron doping also reduces the magnetic ordering temperature T_n of Pr: For the iron-doped samples, $T_N=12$, 7.5, and 5.5 for $x=1$, 0.8, and 0.6, respectively. In the nonsuperconducting quenched samples ($z=6.1$), T_N of the Cu(2) changes little with x: $T_N=415$ K for $x=0$ and 350 K for $x=1$.

The specific-heat measurements which nicely display the Pr magnetic transitions in pure and iron-doped $Y_{1-x}Pr_xBa_2Cu_3O_7$ systems also yield a very high electronic coefficient γ determined from low-temperature measurements. γ increases linearly with increasing x and $d\gamma/dx=3.0(1)$ mJ/mol K^2 in both systems. Since substitution of Cu by Fe also increases γ, the γ values in iron-doped samples are higher than those in the undoped materials. These high γ values are comparable to those of heavy-fermion materials, indicating that the $4f$ level of the Pr ion is strongly hybridized.

II. EXPERIMENTAL DETAILS

The systems $Y_{1-x}Pr_xPr_xBa_2Cu_3O_z$ and $Y_{1-x}Pr_xBa_2Cu_{2.7}Fe_{0.3}O_z$ were prepared by conventional methods.[8] X-ray-diffraction studies were performed to determine the crystallographic structure and the lattice parameters. All systems showed single phase orthorhombic or tetragonal structure; the lattice parameters obtained are given in Table I.

The dc susceptibility measurements of solid ceramic pieces were carried out in a commercial SHE superconducting quantum interference device (SQUID) magnetometer and in a 155 PAR vibrating sample magnetometer in low fields $H \leq 500$ Oe as a function of temperature in the range 2.5–300 K. ac magnetization measurements (1 Oe, 80 Hz) were performed to determine the T_c of the samples, showing results consistent with the dc measurements. The heat capacity was measured over a temperature range of 1.5–70 K in an automated adiabatic calorimeter employing the Nernst step-heating method.

The Mössbauer spectroscopy studies of the ^{57}Fe probe in $Y_{1-x}Pr_xBa_2Cu_{2.7}Fe_{0.3}O_z$ were performed using a conventional constant-acceleration spectrometer and a 50-mCi ^{57}Co:*Rh* source. The spectra at various temperatures were least-squares fitted with several subspectra corresponding to the various inequivalent iron sites.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Crystal structure measurements

X-ray powder diffraction measurements on the $Y_{1-x}Pr_xBa_2Cu_3O_7$ system indicate that all the samples are single phase and have the same orthorhombic structure (Table I). Both a and b lattice parameters increase slightly with x, whereas the c lattice parameter remains practically constant. For the 10 at. % Fe-doped samples the crystal structure is tetragonal, and here also a increases with x (Table I). Surprisingly, no measurable differences were observed in the lattice parameters of $Y_{1-x}Pr_xBa_2Cu_{2.7}Fe_{0.3}O_z$ as a function of z.[10] Based on neutron diffraction studies,[11] we assign the oxygen concentration of iron-doped systems as follows: $z=7.1$ and $z=6.1$ for the fully oxygenated and quenched samples, respectively.

B. Antiferromagnetism of Cu(2) sites determined by Mössbauer spectroscopy

It was shown previously that the Néel temperature of Cu(2) as measured by the Fe probe in $PrBa_2Cu_3O_7$ (325 K) is the same in the samples doped with 1% and 10% Fe,[6] which proves that T_N does not depend on iron concentration; thus iron located in the Cu(2) site is an excellent probe for measuring T_N.[6] In order to determine T_N we used samples in which 10% of the copper was replaced by iron, although iron reduces the T_c of the samples. The samples $Y_{1-x}Pr_xBa_2Cu_{2.7}Fe_{0.3}O_z$, with $z=6.1$ and 7.1, and $x=0.0$, 0.2, 0.4, 0.6, 0.8, and 1.0, have been investigated by ^{57}Fe Mössbauer spectroscopy over a range of temperatures between 90 and 450 K. Some typical spectra obtained at various temperatures and z for $x=0.8$ are shown in Figs. 1 and 2. The well-defined magnetic sextet, which accounts for about 20% of the spectral area, is attributed to iron which replaces copper in the Cu(2) sites and orders antiferromagnetically. As the

TABLE I. Lattice parameters, antiferromagnetic transition temperatures, magnetic data, and electronic specific-heat coefficient for $Y_{1-x}Pr_xBa_2Cu_{3-y}Fe_yO_z$ compounds.

Compound	a (Å)	b (Å)	c (Å)	T_c (K)	T_N (K) for Cu(2)	T_n (K) for Pr	γ (mJ/mol K^2)	χ_0 (emu/mol)	C (emu/mol K)	Θ (K)	P_{eff} (Pr) (μ_B)
$YBa_2Cu_3O_7$	3.825(1)	3.900(1)	11.66(1)	91			17	3.5×10^{-4}	0.14	0.5	
$Y_{0.8}Pr_{0.2}Ba_2Cu_3O_7$	3.830(1)	3.905(1)	11.67(1)	67			64				
$Y_{0.6}Pr_{0.4}Ba_2Cu_3O_7$	3.830(1)	3.921(12)	11.68(1)	35			138				
$Y_{0.4}Pr_{0.6}Ba_2Cu_3O_7$	3.845(1)	3.906(1)	11.69(1)			6.4	180				
$Y_{0.2}Pr_{0.8}Ba_2Cu_3O_7$	3.852(1)	3.916(1)	11.69(1)			12	216				
$PrBa_2Cu_3O_7$	3.873(2)	3.915(1)	11.67(1)			17	300	2.1×10^{-5}	1.22	−8.0	2.97
$YBa_2Cu_{2.7}Fe_{0.3}O_{7.1}$	3.875(1)		11.68(1)	69			73	1.4×10^{-5}	0.40	2.8	
$Y_{0.8}Pr_{0.2}Ba_2Cu_{2.7}Fe_{0.3}O_{7.1}$	3.879(1)		11.69(1)	25			100	1×10^{-3}	0.79	−5.5	3.94
$Y_{0.6}Pr_{0.4}Ba_2Cu_{2.7}Fe_{0.3}O_{7.1}$	3.881(1)		11.70(1)		30(3)		225	1.23×10^{-3}	0.89	−5.2	3.13
$Y_{0.4}Pr_{0.6}Ba_2Cu_{2.7}Fe_{0.3}O_{7.1}$	3.884(2)		11.70(1)		230(10)	5.5	270	1.13×10^{-3}	1.09	−9.7	3.03
$Y_{0.2}Pr_{0.8}Ba_2Cu_{2.7}Fe_{0.3}O_{7.1}$	3.909(1)		11.73(1)		300(10)	7.5	280	1.27×10^{-3}	1.25	−8.6	2.92
$PrBa_2Cu_{2.7}Fe_{0.3}O_{7.1}$	3.917(1)		11.75(1)		325(10)	12	340				

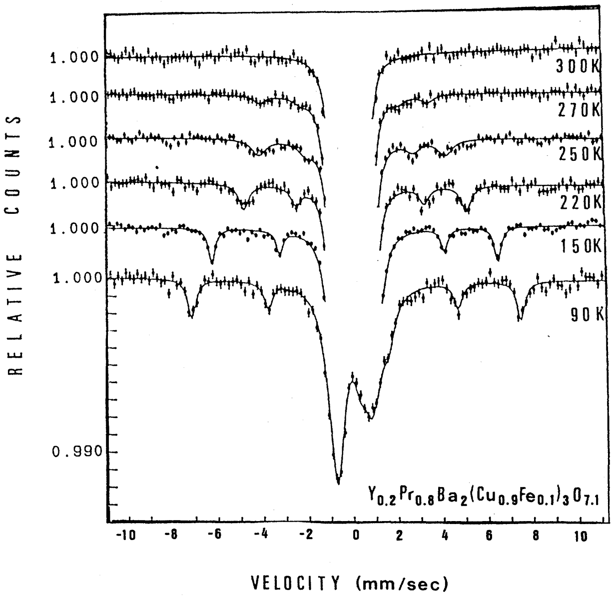

FIG. 1. Mössbauer spectra of ^{57}Fe in $Y_{0.2}Pr_{0.8}Ba_2(Cu_{0.9}Fe_{0.1})_3O_{7.1}$ at several temperatures.

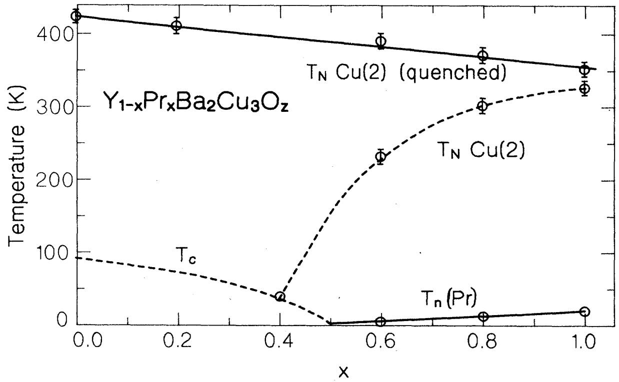

FIG. 3. Superconducting-magnetic phase diagram for $Y_{1-x}Pr_xBa_2Cu_3O_7$ system. The antiferromagnetic transition temperatures for the Cu(2) sites in both full oxygen and quenched samples were obtained with 10 at. % a Fe-doped samples.

temperature is raised, the magnetic splitting decreases and the Néel temperature in each of the compounds can be obtained. In Fig. 3 the change of T_N as a function of x and z is displayed. Numerical values for the $z=7.1$ system are given in Table I. The T_N value for $x=0.4$ was obtained by direct magnetization measurements (Fig. 4) to be discussed in the following. T_N decreases gradually to 230 K for $x=0.6$, then drops sharply to below 35 K for $x=0.4$. Mössbauer measurements at 4.1 K also show that at this temperature the Cu(2) site is magnetically ordered. This could presumably point to coexistence between superconductivity and magnetic order for this compound. However, it is possible that the presence of iron may change T_N slightly. It is also possible that due to slight inhomogeneity of the Pr content in the sample, there are regions with $x>0.4$ in which Cu(2) is magnetically ordered and regions with $x<0.4$ which are superconducting. It is thus impossible to make any definite conclusions about the coexistence of superconductivity and magnetic order.

In order to compare the ordering temperature of Cu(2) in oxygen-rich and oxygen-deficient samples,[2] all the iron-doped samples listed in Table I were quenched from 900 °C to liquid nitrogen temperature. Surprisingly, no measurable differences were observed between the lattice parameters of the quenched and the oxygen-rich samples. The magnetic-ordering temperatures of the Cu(2) site in the $Y_{1-x}Pr_xBa_2Cu_{2.7}Fe_{0.3}O_{6.1}$ system, measured in the same manner as for $z=7.1$, range from 350 K for $x=1$ to 415 for $x=0$, and are also shown in Fig. 3.

C. dc and ac magnetization measurements

The variation of T_c in the $Y_{1-x}Pr_xBa_2Cu_3O_7$ system was reported recently by several authors.[4,5] Our data obtained by ac and dc susceptibility measurements are ex-

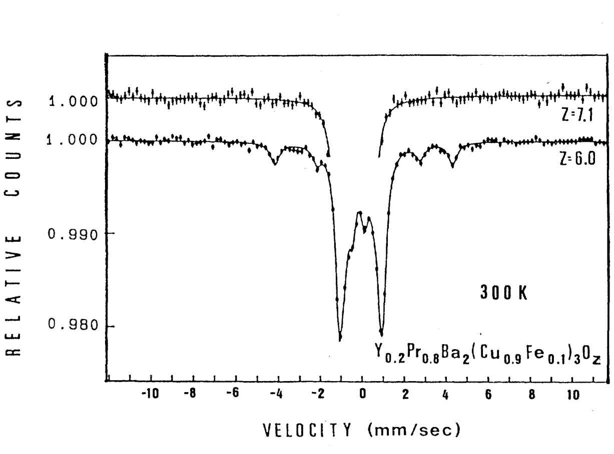

FIG. 2. Mössbauer spectra of ^{57}Fe in $Y_{0.2}Pr_{0.8}Ba_2(Cu_{0.9}Fe_{0.1})_3O_z$ at 300 K. Note that the spectra for the quenched ($z=6.1$) sample show a magnetic subspectrum.

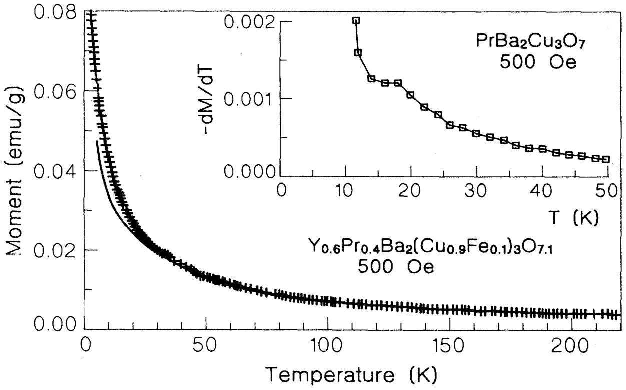

FIG. 4. The temperature dependence of the magnetization at 500 Oe for $Y_{0.6}Pr_{0.4}Ba_2(Cu_{0.9}Fe_{0.1})_3O_{7.1}$. The solid line is the Curie-Weiss theoretical curve. The inset shows the temperature derivative of the susceptibility for $PrBa_2Cu_3O_7$.

hibited in Fig. 3, and are in good agreement with the published results. Clearly, T_c decreases with increasing x, and for $x>0.55$ the compounds are not superconducting. The valence of Pr in this system is controversial. The magnetic susceptibility measurements reveal that the effective magnetic moment corresponds to an average valence of 3.87 for Pr in the entire range of x.[4] On the other hand, soft x-ray absorption spectroscopy indicates that the Pr ions are trivialent for all values of x.[5] It is possible that Pr^{3+} is found only at the surface, while in the bulk the Pr valence is higher. The effective paramagnetic moments according to Hund's rule for Pr^{3+} and Pr^{4+} are $3.58\mu_B$ and $2.56\mu_B$, respectively. The temperature dependence of the susceptibility measured at 500 Oe by the SQUID magnetometer is found to be well characterized by the Curie-Weiss law, $\chi=\chi_0+C/(T-\Theta)$; the values for the 10% iron-doped samples given in Table I were calculated by the least-squares method. For $0<x\leq 0.6$ calculations were made for a temperature range above T_N of Cu(2); for $x=0.8$ and 1.0 the temperature range chosen was between 50 and 280 K. To obtain the paramagnetic moment for "pure" Pr given in Table I, we subtracted the Curie constants for $YBa_2Cu_{2.7}Fe_{0.3}O_{7.1}$ (0.4 emu/mol) from the C values obtained. The P_{eff} obtained, 3.0(1) (except for $x=0.2$), is a clear indication that Pr is in a mixed-valent state with an average formal valence of about 3.6. The Pr valence obtained here agrees well with that reported previously on an undoped Fe system, namely 3.67.[12] As seen from the values of χ_0 and P_{eff} given in Table I, it is evident that their values are not affected by whether the Cu(2) site is ordered or not.

For the $x=0.8$ and $x=0.6$ samples, the curves calculated according to Curie-Weiss law deviate from the experimental data at T_N, the temperature obtained as the ordering temperature of Cu(2) by Mössbauer spectroscopy. This deviation occurs for $x=0.4$ at about 30 K, which is therefore assumed to be T_N for this sample.

Several mechanisms for the absence of superconductivity in pure $PrBa_2Cu_3O_7$ and for the suppression of T_c in $Y_{1-x}Pr_xBa_2Cu_3O_7$ with increasing x are suggested. One possible mechanism is associated with the fact that Pr is in an intermediate valence state. The presence of mixed valent Pr, instead of Y, midway between the Cu-O planes reduces the d-hole concentration in the plane. It would seem reasonable to suggest another possible mechanism, based on a magnetic pair-breaking effect. However, the latter can be excluded, since in $Y_{1-x}Gd_xBa_2Cu_3O_7$ no detrimental effect on T_c has been observed, even though Gd has a much larger spin than Pr.[13] However, a possible mechanism which combines the two above-mentioned mechanisms is based on the fact that Pr is the largest rare-earth ion having $4f$ electrons which forms the orthorhombic $RBa_2Cu_3O_7$ structure. When a small Y^{3+} ion (0.93 Å) is replaced by a larger Pr^{3+} ion (1.09 Å) there is an increase of about 17% in the ionic radius of R, yet the orthorhombic unit cell parameters scarcely change. The Pr $4f$ electrons hybridize with the hole carriers of the Cu(2) planes, so that the reduction in T_c may be due to the weakening of the superconducting pairs through the magnetic pair-breaking effect. This hybridization also changes the effective number of Pr $4f$ electrons (mixed valence state) which leads to the reduced magnetic moment observed for the whole range of Pr concentrations. On the other hand, the exchange interactions Pr-Pr are strengthened, and lead to a high magnetic-ordering temperature in the Pr sublattice. The observed T_N is almost 2 orders of magnitude higher than theoretically expected when compared to the ordering temperature of Gd. Once superconductivity disappears, the Cu(2) sites also become antiferromagnetically ordered as in the $YBa_2Cu_3O_z$ system for $z>6.5$.

D. Determination of T_n for Pr

To determine the ordering temperature of Pr in $Y_{1-x}Pr_xBa_2Cu_3O_7$, susceptibility measurements were made at low temperatures in various applied fields. The general behavior differs from what is expected based on a simple antiferromagnetic structure. In all cases, the temperature dependence of the susceptibility monotonically increases with decreasing temperature (Fig. 4). For $x=1$ we observed a pronounced change in the slope of the magnetization curve at T_n (inset of Fig. 4). For $x<1.0$ it is not possible to determine T_n unequivocally from magnetization measurements alone. This could be due to the small magnetic moment observed for Pr ($0.74\mu_B$) by neutron diffraction measurements,[7] or it may indicate that the magnetic structure is more complicated than the original antiferromagnetic model. On the other hand, the specific-heat measurements clearly show an anomaly in the vicinity of T_n.

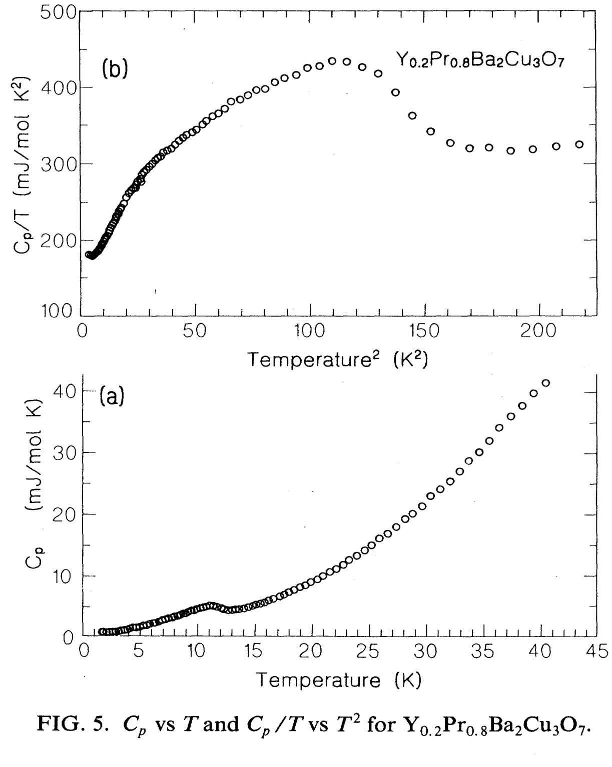

FIG. 5. C_p vs T and C_p/T vs T^2 for $Y_{0.2}Pr_{0.8}Ba_2Cu_3O_7$.

The specific heat of $Y_{0.2}Pr_{0.8}Ba_2Cu_3O_7$ over the temperature range of 5–40 K is shown in Fig. 5. The usual presentation of C_p/T versus T^2 at low temperatures is also shown. The anomaly observed at 12 K is obvious. The magnetic entropy associated with this transition is 1.2 J/mol K. For $PrBa_2Cu_{2.7}Fe_{0.3}O_{7.1}$ the entropy obtained for the magnetic transition is 1.1 J/mol K. These values are much lower than the 5 J/mol K obtained by Li *et al.* for $PrBa_3Cu_3O_7$.[7] In this respect it should be noted that a peak in the specific heat was observed at about 19 K by Sankar *et al.*,[14] but it was interpreted as due to Schottky anomalies of the Pr ions, which is a completely different interpretation than the one presented here.

Figures 6 and 7 exhibit the C/T versus T^2 curves for all samples measured. One observes in these figures that in both systems T_n for Pr ions decreases with x. Values obtained for both systems are given in Table I. In neither system is a magnetic-order transition observed for $x=0.4$ down to 2.5 K. The deviation from linearity for $x=0.4$ in the iron-doped sample is probably due to inhomogeneity of the sample. Note that T_n is lowered by the presence of Fe, probably due to some frustration caused by the strongly magnetic iron. The entropy associated with the magnetic transition for $x=0.6$ in undoped and Fe-doped samples is 0.4 and 0.36 J/mol K, respectively. At very low temperatures the strong upturn in C/T, common in such materials, is observed in both systems. The electronic specific-heat coefficient γ was calculated by the usual procedure of extrapolating the nearly linear variation of the C/T values from above T_n to $T=0$. The data obtained are given in Table I. All samples measured showed very large γ values, characteristic of "heavy-fermion" systems. γ increases as a function of x, with a slope of $d\gamma/dx=3.0$ mJ/mol K^2. The change in γ does not depend on whether the samples are superconductors or not. Substitution of Cu by 10% at Fe in $YBa_2Cu_3O_7$ significantly increases γ (to 73 mJ/mol K^2). This appears to be a systematic trend in $YBa_2Cu_3O_7$ when Cu is replaced by either Zn or Ni. An analogous trend is also observed in $La_{1.85}Sr_{0.5}CuO_4$ when Cu is replaced by Zn.[15] $d\gamma/dx$ obtained here is much larger than the $d\gamma/dx=2.1$ mJ/mol Cu K^2 obtained when Cu is replaced by other transition metals, and in fair agreement with data obtained in Ref. 16. The same slope in γ is obtained in the 10 at. % Fe-doped samples, but the values are higher due to the Fe contribution to γ.

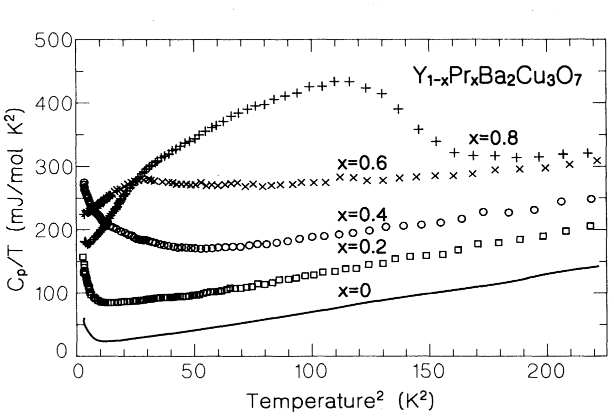

FIG. 6. C_p/T vs T^2 curves for $Y_{1-x}Pr_xBa_2Cu_3O_7$ at low temperatures.

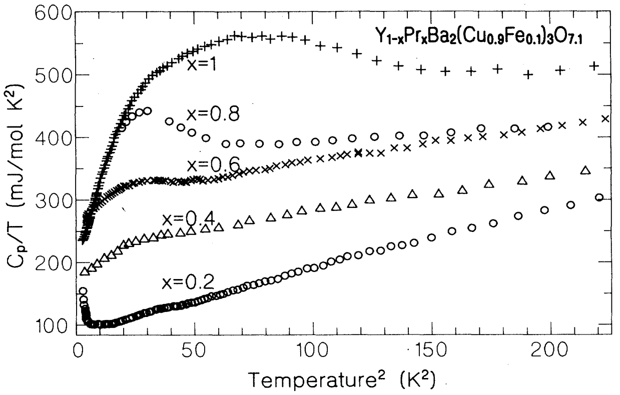

FIG. 7. C_p/T vs T^2 curves for $Y_{1-x}Pr_xBa_2(Cu_{0.9}Fe_{0.1})_3O_{7.1}$ at low temperatures.

High γ and χ_0 values are characteristic of heavy-fermion systems. The γ/χ_0 ratio is enhanced significantly when Pr is substituted for Y (see Table I). The heavy-fermion behavior of the $Y_xPr_{1-x}Ba_2Cu_3O_7$ system is probably associated with the mixed-valent character of Pr. Thus the mixed-valent state for Pr affects not only the electronic states of the system, but also the superconducting properties of the Cu(2)-O layers (suppression of T_c) and the magnetic interactions in both Cu(2) and Pr sites. This allows Pr ions to interact with each other via mechanisms mediated by conduction electrons, i.e., not via dipolar interaction as appears to be the case for other rare-earth elements.

IV. CONCLUSION

Based on the studies reported here we can summarize the behavior of the $Y_{1-x}Pr_xBa_2Cu_{3-y}Fe_yO_z$ systems as follows.

(a) For $z=7$ the system is superconducting (below T_c) for low x values, but antiferromagnetic at the Cu(2) site (below T_N) and the Pr site (below T_n) for high values of x.

(b) There is no unambiguous observable evidence for an overlap of superconductivity and antiferromagnetism.

(c) For $z=6$ the Cu(2) site is antiferromagnetic for all x values, and T_N varies little with x: T_N $(x=0)=415$ K and T_N $(x=1)=350$ K.

(d) The effect of the substitution of Fe for Cu is to reduce T_c and the magnetic-ordering temperature T_n of Pr.

(e) The electronic specific-heat coefficient γ increases linearly with x, regardless of whether the materials are superconducting or magnetic. In this respect the systems behave as heavy-fermion systems.

All the phenomena reported are explained by assuming the hybridization of the extended Pr $4f$ electron orbitals with the electrons (holes) of the Cu(2)-O layers.

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