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Superconductivity of the Pr-doped (Y,Eu)$Ba_2Cu_4O_8$ system

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A systematic study of the T_c dependence of the disordered $R_{1-x}Pr_xBa_2Cu_4O_8$ (RPr-124) system (R=Y,Eu) on the Pr concentration x is reported here. Like $(Y,Eu)_{1-x}Pr_xBa_2Cu_3O_7$ (RPr-123), T_c of these systems decreases monotonically with Pr concentration. dT_c/dx, however, is found to be smaller in RPr-124 than in RPr-123. Moreover, unlike RPr-123, dT_c/dx does not vary monotonically with x at least in the YPr-124 system, although the relative variation of T_c with the rare-earth (Y,Eu) ionic radius is similar in both the cases. The observed results are discussed in terms of the hole-filling model and the Abrikosov-Gorkov (AG) theory. It is found that the AG theory is not adequate to explain the observed T_c variation with x in the RPr-124 systems. We have attributed the failure of the AG theory in the RPr-124 systems to the linear x dependence of the carrier-impurity scattering rate. A modified AG theory considering the observed carrier-impurity scattering rate is found to provide a reasonable explanation of T_c variation in the RPr-124 systems. The different T_c variations in the YPr-124 and EuPr-124 systems are qualitatively understood on the basis of an indirect effect of the Pr impurity on the coupling of the Y,Eu state with the Cu 3d–O 2p state. The relatively smaller value of dT_c/dx in Y,Eu-124 than in Y,Eu-123 is understood on the basis of the larger separation of the CuO_2-(Y,Eu/Pr)-CuO_2 regions in the former system.

I. INTRODUCTION

Of all the rare-earth-based cuprate superconductors with the orthorhombic 1-2-3 structure. $PrBa_2Cu_3O_7$ possesses a special stature. Unlike other rare-earth-based cuprates it is nonsuperconducting and highly semiconducting even though it possesses identical crystallographic structure to superconducting cuprates.[1,2] The two other exceptions, based on Tb and Ce, do not result in the formation of the orthorhombic structure; they only lead to the formation of mixed phases.[3]

The superconducting transition temperature T_c of the system $R_{1-x}Pr_xBa_2Cu_3O_7$ has been seen to decrease with Pr content x and eventually superconductivity disappears at a critical concentration x_{cr} varying from 0.32 in the SmPr-123 system to 0.60 in the TmPr-123 system depending on the rare-earth ionic radius.[4,5] Different studies[2,6,7] of the Hall effect, magnetic susceptibility, etc., suggest Pr to exist in a higher than 3+ oxidation state in these materials, the extra electron compensating a hole in the Cu-O network and thereby decreasing the T_c. Since the band filling due to Pr^{4+} is expected to be nearly the same in different rare-earth-containing systems, this alone cannot explain the strong x_{cr} dependence observed for these systems. Spectroscopic studies,[8–10] on the other hand, suggest the hybridization of the Pr 4f states with Cu 3d–O 2p states, based on a nearly Pr^{3+} valence state. Such a contention also receives credence from the relatively high values of electronic specific-heat coefficients and the magnetic ordering temperature of Pr moments in the Pr-123 system.[11–13] The results have been reasonably well understood on the basis of the Abrikosov-Gorkov (AG) theory of pair breaking due to spin-dependent exchange scattering of mobile holes in the Cu-O valence band by magnetic Pr ions.[14–16]

While considerable efforts have been made to understand the effects of Pr incorporation in R-123, there are only a few reports[17–19] about the superconducting behavior of the Pr-substituted R-124 system possessing double Cu-O chain networks. It would be worthwhile to study the Pr-doped R-124 system, and to examine the applicability of the hybridization and the pair-breaking concepts (AG theory).

We report here a systematic study of the T_c variation as a function of Pr concentration in R-124. An attempt has been made to explain the different x_{cr} concentrations and dT_c/dx observed for different rare earths (R =Y and Eu) on the basis of a modified AG theory.

II. EXPERIMENTS

A series of samples of $Y_{1-x}Pr_xBa_2Cu_4O_8$ (YPr-124) and $Eu_{1-x}Pr_xBa_2Cu_4O_8$ (EuPr-124) with $0 \le x \le 1.0$ has been synthesized by the solid-state reaction process utilizing the appropriate quantities of the various ingredients Y_2O_3, Eu_2O_3, $Ba(NO_3)_2$, CuO, and Pr_6O_{11} having purity better than 99.99%. High-pressure oxygen treatment to synthesize the 1-2-4 compound was circumvented by using a small quantity of $Na_2C_2O_4$ (0.2 mol) as catalyst. The pulverized mixture of the oxides and the catalyst was heat treated at 900 °C for 12 h in an O_2 environment at ambient pressure. The second treatment was given at 820 °C for 48 h, again in flowing oxygen, with final quenching to ambient temperature. Details of the sample preparation are provided elsewhere.[20]

The samples were examined through x-ray diffractometry for homogeneity and phase formation using a Siemens D-500 diffractometer with Cu $K\alpha$ radiation. The conventional four-probe technique was employed to obtain the resistivity behavior in the tempera-

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ture range of 4.2–300 K. The temperature of the samples was monitored using a standard platinum resistance thermometer of 100 Ω in conjunction with a Keithley 195A digital multimeter with an accuracy of ±0.1 K in the range of 70–300 K. A liquid-helium cryostat with Ge sensor was used in the range 4.2–70 K. The entire system, comprising digital nanovoltmeter, constant-current source, and the temperature controller and indicator, was hooked to an IBM PC for automatic data acquisition and control.

III. RESULTS

Typical x-ray diffractograms of YPr-124 and EuPr-124 samples are shown in Figs. 1(a) and 1(b), respectively. The orthorhombicity of the pristine system decreases with increasing Pr concentration. Lattice parameters a_0, b_0, and c_0 of the undoped Y-124 system were found to be 3.83, 3.87, and 27.16 Å, respectively. The parameter c_0 of the various Pr-doped samples was found to be the same as that of the undoped sample and is in good accord with the reported data.[3] Variation of the lattice parameters with Pr doping in R-124 seems to follow the same trend as in the RPr-123 system.[3,5,9]

Resistivity-temperature data for the YPr-124 series ($0 \leq x \leq 0.9$) are presented in Fig. 2. Y-124 in its normal state has a highly metallic character with an extrapolated negative intercept at the resistivity axis, indicating the good quality of the starting material.[20] As Pr content x

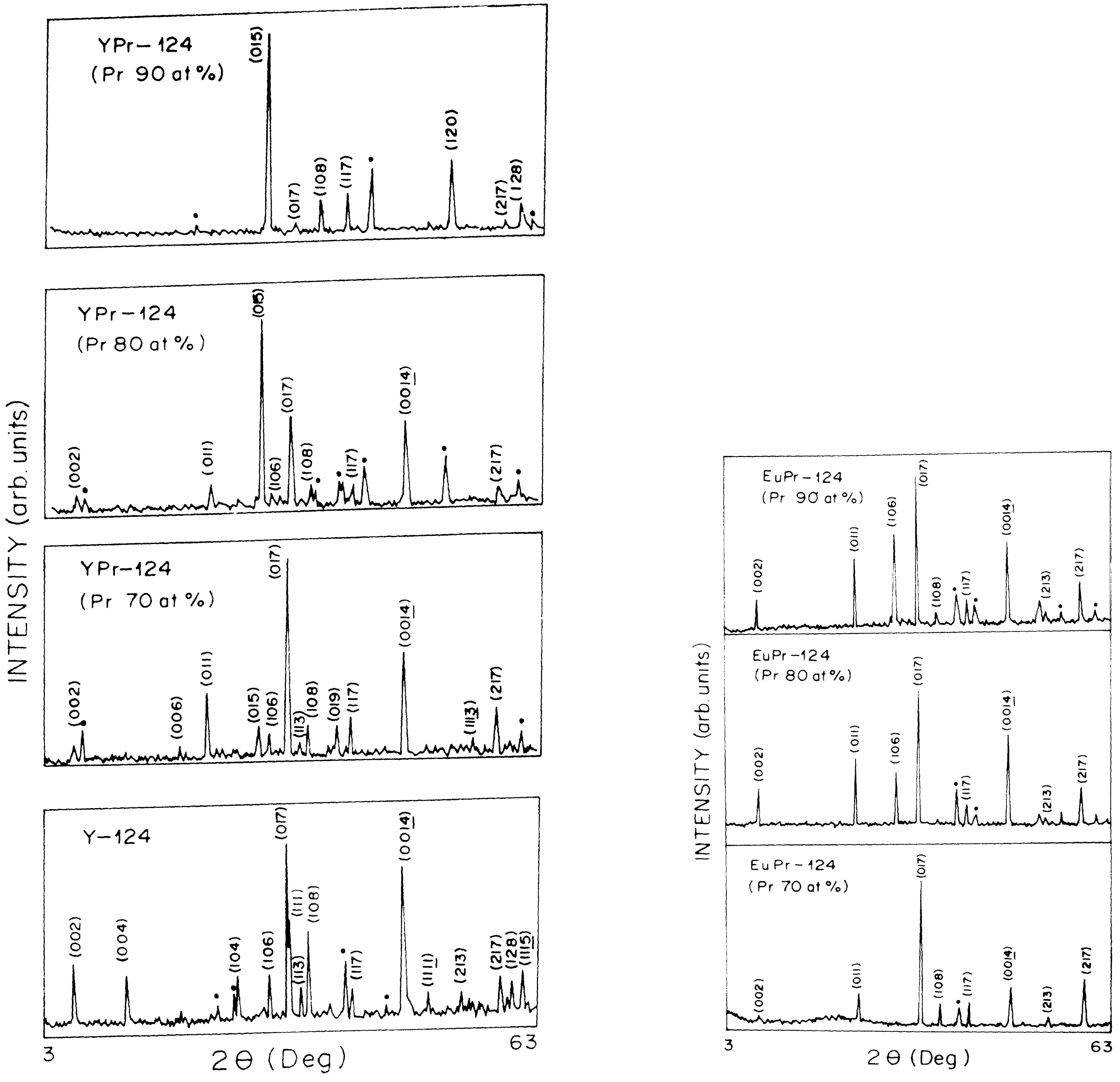

FIG. 1. (a) X-ray diffractograms for the $Y_{1-x}Pr_xBa_2Cu_4O_8$ samples. (b) X-ray diffractograms for the $Eu_{1-x}Pr_xBa_2Cu_4O_8$ samples.

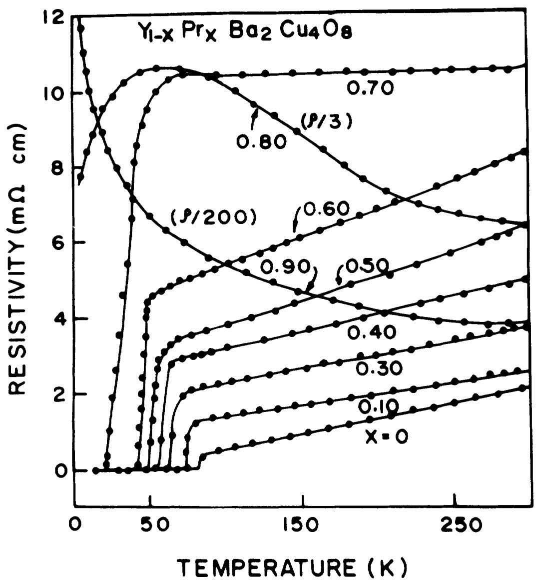

FIG. 2. Resistivity vs temperature plot of the $Y_{1-x}Pr_xBa_2Cu_4O_8$ system.

increases, a crossover from metallic to semiconducting-like behavior is seen around $x=0.70$. Though such a trend of the normal-state resistivity has been reported in the RPr-123 system, the T_c depression rate is much lower in the (*R*Pr)-124 system.[17,18] Here even the doped material with 60 at. % Pr superconducts at 37 K while in (RPr) 123 50 at. % Pr doping renders the material semiconducting.[3,9] The results obtained on EuPr-124 (Fig. 3) are essentially similar to those of YPr-124. The Eu-124 sample has a lower $T_c(R=0)$ of ~70 K compared to 80 K for Y-124. Also the critical concentration (x_{cr}) of Pr in Eu-124 is much lower ($x_{cr}=0.60$) as compared to that in Y-124 ($x_{cr}=0.80$). T_c depression with x is seen to be much stronger in EuPr-124 than in YPr-124 (cf. Fig. 4).

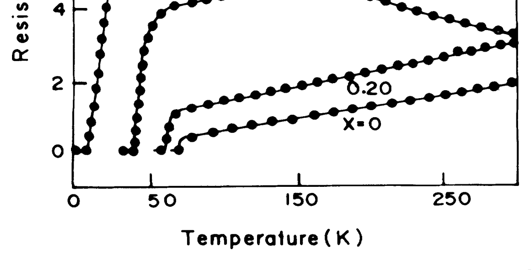

FIG. 3. Resistivity vs temperature plot of the $Eu_{1-x}Pr_xBa_2Cu_4O_8$ system.

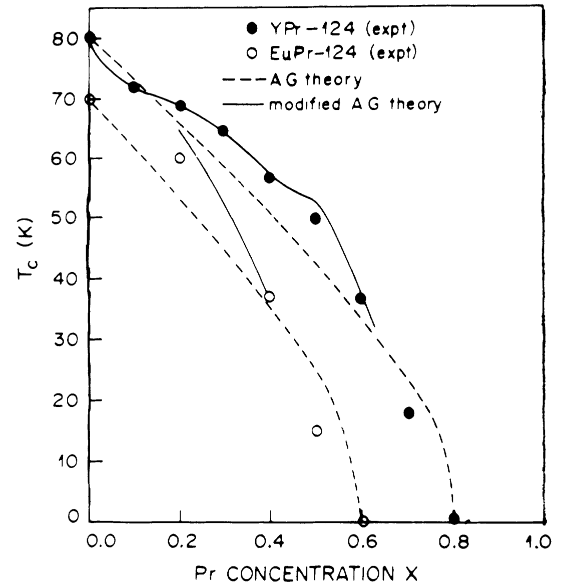

FIG. 4. Transition temperature T_c vs Pr concentration x for $Y_{1-x}Pr_xBa_2Cu_4O_8$ and $Eu_{1-x}Pr_xBa_2Cu_4O_8$ systems. Solid circles (YPr-124) and open circles (EuPr-124) show the observed values. Dashed lines show the result of AG theory. Solid lines show the result of the modified AG theory.

IV. DISCUSSION

The observed results suggest a subtle difference in the T_c variation of the YPr-124 and EuPr-124 systems with x. T_c depends not only on the presence of Pr ions but also on the nature of the Eu and Y ions, which is borne out by the fact that the critical concentration x_{cr} of Pr is different for the YPr-124 ($x_{cr}=0.80$) and EuPr-124 ($x_{cr}=0.60$) systems. These values are significantly higher than the 0.67 and 0.45 found for the corresponding YPr-123 and EuPr-123 systems, respectively.[5] This apart, the curvature of the T_c-x changes in the YPr-124 system unlike in the YPr-123 system. The data on the EuPr-124 system, however, are not sufficient to show whether there is any T_c-x curvature change in this system or not.

On the basis of the above results it is clear that T_c variation in the disordered 1-2-4 systems is much more complex than in the 1-2-3 systems. While the relative variation of T_c with x in the Y- and Eu-based 1-2-4 systems,

$$(dT_c/dx)_{\text{Y-124}}-(dT_c/dx)_{\text{Eu-124}} ,$$

can be understood on the basis of the different ionic radii of the Y and Eu ions in the system, we need to invoke other ideas to understand the difference in the T_c variation between 1-2-3 and 1-2-4 systems for a given rare-earth ion Eu or Y. This is because the two models, viz., hole-filling phenomenology and the Abrikosov-Gorkov pair-breaking theory, which are used to explain the T_c

variation, are not separately valid in the present case, as both these ideas lead to the monotonic variation of (dT_c/dx) with x which is not the case for the 1-2-4 system. In the following we attempt to establish a relation between the observed T_c depression and the AG theory and the hole-filling model in a phenomenological manner. In the AG theory the T_c variation as a function of concentration x is given by[14]

$$\ln\frac{T_c}{T_{c0}}=\psi(\tfrac{1}{2})-\psi\left[\frac{1}{2}+\frac{1}{2\pi\tau_{\mathrm{AG}}T_c}\right], \tag{1}$$

where T_{c0} is T_c for $x=0$, ψ is the psi or digamma function, and τ_{AG}, the scattering time in the AG theory, is given by

$$\frac{1}{\tau_{\mathrm{AG}}}=2\pi x N_F J_{\mathrm{SF}}^2(g_J-1)^2 J(J+1)\ . \tag{2}$$

Here N_F is the density of states at the Fermi level, J_{SF} is the parameter for exchange interaction $-2J_{\mathrm{SF}}\mathbf{S}\cdot\mathbf{s}$ due to the interaction of carrier spin $\mathbf{s}$ with Pr spin $\mathbf{S}$, g_J is the Landé g factor, and J is the total angular momentum of the Pr ions. It is worth noticing that while the factor $x(g_J-1)^2J(J+1)$ depends solely on the Pr ions, $N_F J_{\mathrm{SF}}^2$ depends on both the Pr ions and the Y- or Eu-based system. Based on this theory, we calculate T_c for different impurity concentrations x. We have used those values of τ_{AG} which give $T_c=0$ at the observed values of $x_{\mathrm{cr}}=0.80$ for YPr-124 and $x_{\mathrm{cr}}=0.60$ for EuPr-124. x_{cr} is given by[14]

$$x_{\mathrm{cr}}=2\gamma/\pi T_{c0}\ , \tag{3}$$

where $\gamma=\exp[(-\psi(1)]$ is the Euler constant. We could have taken those values of τ_{AG} for which $(dT_c/dx)_{x=0}$ is given by the AG theory as in Ref. 4, but then the values of x_{cr} would have been different from the observed values. Ideally, for any successful theory both these approaches should lead to the same values of τ_{AG} and x_{cr}.

Within the framework of the AG theory the observed value of x_{cr} requires that in

$$\frac{1}{\tau_{\mathrm{AG}}}=(n_0e^2/m)\rho_{\mathrm{AG}}(x)=(n_0e^2/m)x\rho'_{\mathrm{AG}} \tag{4}$$

$\rho'_{\mathrm{AG}}=7.0$ (8.9) mΩ cm/Pr concentration in the YPr-124 (EuPr-124) system. In Eq. (4), ρ_{AG} is the resistivity corresponding to the scattering rate $1/\tau_{\mathrm{AG}}$. n_0, m, and e are carrier density, carrier mass, and electronic charge, respectively. The results of our calculations are shown in Fig. 4 by dashed lines. It is also seen from this figure that the AG theory based on the observed x_{cr} values, as mentioned above, is not able to explain the dT_c/dx behavior in RPr-124 systems. In fact, the calculated T_c in the case of YPr-124 is lower in comparison to the observed T_c for $0.15\le x\le 0.65$ and is higher for $x\le 0.15$ and $x\ge 0.65$. Similar qualitative differences remain for the EuPr-124 system for $x\gtrsim 0.45$. It is clear from Fig. 4 that the AG theory provides only a monotonic dT_c/dx variation for $0\le x\le x_{\mathrm{cr}}$, unlike the observed behavior which changes curvature below $x\approx 0.2$ and above $x\approx 0.6$ for YPr-124 and at $x\approx 0.4$ for EuPr-124. Such a feature is apparently absent in the RPr-123 system.[7]

A. Modified AG theory

For a reasonable understanding of the observed data (Fig. 4) we consider a suitable modification of the AG theory. The AG theory is based on a clean system described by the Bardeen-Cooper-Schrieffer (BCS) theory where the pairing is of s-wave type. On the basis of the observed gap ratio[21] and penetration depth[22] of the clean and dirty Y-124 systems, the pairing in this system seems to be of s-wave type. There is, however, another important condition for the validity of the AG theory, viz., the situation of the existing interaction should be such that the scattering rate (or equivalently the resistivity) due to impurity is proportional to the impurity concentration. Let $\rho_0(x)$ denote the resistivity due to impurity. It is given by $\rho_0(x)=\rho(x,T=0)$ where $\rho(x,T)$ is the resistivity shown in Figs. 2 and 3. In Fig. 5 we have plotted $\rho_0(x)$ with x. It is clear that $\rho_0(x)$ is not proportional to x. Thus we cannot expect AG theory to be applicable in such a situation.

A phenomenological modification of the AG theory is obtained when $\rho_{\mathrm{AG}}(x)$ [cf. Eq. (4)] is replaced by $\rho_0(x)$. The scattering rate will then not vary linearly with x. Let $1/\tau_0$ denote the scattering rate corresponding to the resistivity $\rho_0(x)$. Then

$$1/\tau_0=(n_0e^2/m)\rho_0(x)\ . \tag{5}$$

We may express the modification caused by the replacement of $\rho_{\mathrm{AG}}(x)$ by $\rho_0(x)$ in terms of an effective impurity concentration x_{eff} by requiring that

$$1/\tau_0=(n_0e^2/m)x_{\mathrm{eff}}\rho'_{\mathrm{AG}}\ . \tag{6}$$

Comparing Eqs. (5) and (6) we obtain

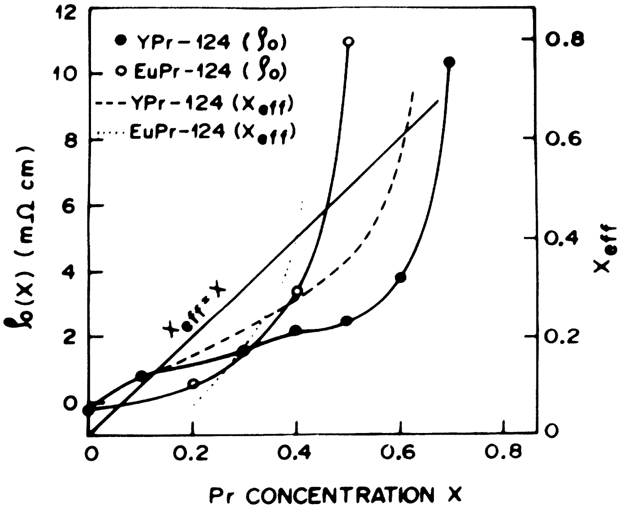

FIG. 5. Observed $\rho_0(x)$ vs x for $Y_{1-x}Pr_xBa_2Cu_4O_8$ and $Eu_{1-x}Pr_xBa_2Cu_4O_8$ systems. Solid lines are guides to the eye. The dashed (dotted) lines show the values of x_{eff} for YPr-124 (EuPr-124). The $x_{\mathrm{eff}}=x$ line is also shown.

$$x_{\text{eff}}=\rho_0(x)/\rho'_{\text{AG}} \ . \tag{7}$$

Values of x_{eff} are plotted in Fig. 5. T_c values as calculated by the modified AG theory in accordance with the scattering rate given by Eq. (6) are shown in Fig. 4 by solid lines. Since we have used the observed values of $\rho_0(x)$ in these calculations it is not possible to obtain T_c for all x values as exhibited in Fig. 4. An excellent agreement for the T_c depression is seen in the case of the YPr-124 system while in the EuPr-124 system it is not fully conclusive due to lack of sufficient data. More work along these lines is in progress.

The fact that up to $x=0.6$ the modified AG theory provides a satisfactory explanation of the T_c depression in the YPr-124 system implies that up to this Pr concentration the carrier-impurity scattering is mainly magnetic in nature. Beyond $x=0.5$ the modified AG theory predicts too rapid a T_c depression to fall in line with the observed behavior. Certainly the Pr impurities have started to cause strong potential scattering after $x=0.6$. In fact in the YPr-123 system it is believed that the Pr $4f$ orbitals hybridize with the Cu $3d$–O $2p$ valence state, resulting in an effective Pr valence[7] of 3.3. Recent theoretical calculations by Fehrenbacher and Rice[23] on the basis of a linear combination of Pr^{4+} and Pr^{3+} plus a ligand hole state show a Pr valence of 3.15–3.20. The significance of such a hybridization is also clear from the fact that Zn doping in YPr-123 changes the magnetic order from that in the Zn-free system.[24]

The success of the modified AG theory in the YPr-124 system suggests the nature of the pairing to be of s-wave type. This seems to contradict NMR observations on Zn-doped Y-123 system.[25] These observations, however, can also be understood in terms of s-wave pairing. The point is that, if the impurities suppress the pairing interaction,[26] then T_c will decrease significantly with s-wave pairing also. In this sense the present modified theory has a more general relevance in the cuprate superconductors than envisaged earlier.

B. Effect of Y and Eu

In the presence of Pr atoms the charge transfer from the rare-earth sites (Y,Eu) to the CuO_2 layers in the 1-2-3 and 1-2-4 systems will be affected. In fact the Pr $4f$–(Cu $3d$–O $2p$) hybridization will have an induced effect on Y and Eu also. In the Y-124 system $4d$ states of Y atoms will participate in the hybridization process while in Eu-124 $4f$ states will take part. The energy difference between the Y^{3+} and Y^{2+} states is expected to be larger than that of the corresponding Eu^{3+} and Eu^{2+} states. This is because the $4d$ state (of Y^{2+}) is completely empty in the case of Y^{3+}, unlike the case of Eu^{2+} and Eu^{3+} where the $4f$ state is partly filled in both valence states.

Because of the larger energy difference in the case of Y, its states are expected to have relatively less hybridization with the Cu $3d$–O $2p$ states due to the induced effect of the Pr atoms. The smaller ionic size of Y^{3+} also favors this result (see above). The main effect of this behavior will be that in the YPr-124 system Y^{3+} ions will remain relatively unaffected and the source of the carrier self-energy will be only the Pr ions. On the other hand, in the EuPr-124 system, the Eu ions will be affected by the Pr ions and the carrier self-energy will be enhanced in comparison to the YPr-124 system, resulting in a faster T_c depression.

Thus the difference in the T_c depression rates in the YPr-124 and EuPr-124 systems is due to an indirect disorder effect on Y and Eu sites by the Pr ions. According to Fig. 5, for $x<0.3$, Eu atoms in EuPr-124 do not let the Pr atoms reduce the carrier concentration in the CuO_2 layers. This is possible in two ways. First, the Pr atoms may interact directly with the Eu atoms rather than affecting the carrier density, and secondly (for $x<0.3$) Eu atoms compensate the loss of carrier concentration indirectly. For $x>0.3$, the role of Eu atoms is reversed. In reality, the effects of Y and Eu, as revealed by Fig. 5, are quite complex and more data on Eu- as well as on other rare-earth-based 1-2-4 systems need to be generated. Such a study is already in progress.

C. Comparison with the (Y,Eu)-123 system

In order to see why (Y,Eu)Pr-124 and (Y,Eu)Pr-123 systems show different behavior of the superconductivity, we proceed as follows. There are, in general, four different effects in disordered high-T_c systems:

(a) change in the pairing interaction V,
(b) change in the Coulomb interaction U^*,
(c) appearance of normal self-energy, and
(d) inelastic scattering effects leading to reduction in T_c.

While the effects (a), (b), and (c) will appear in disordered systems only, the effect (d) will be present in both ordered and disordered systems. In the AG theory, it is the appearance of normal self-energy (c) which is considered to lower T_c in disordered systems. The other effects are not taken into account. The effects (a), (b), and (c) have been studied in conventional superconductors[27] where the pairing mechanism is based on the electron-phonon interaction. Disorder is seen to increase both the pairing interaction and the Coulomb interaction. If the increment in the values of the pairing and Coulomb interactions is exactly the same for all x, then only the normal self-energy will determine the T_c variation, making the AG theory applicable in such a situation.

Owing to differences in the crystal structures of R-123 and R-124, it seems natural to expect different variations in the pairing and Coulomb interactions in these systems. For some impurity concentrations pairing interaction may dominate over the Coulomb interaction, and vice versa for other concentrations.

Let us now examine the higher x_{cr} value observed in YPr-123 or YPr-124 as compared to that in EuPr-123 or EuPr-124. There exist mainly two interactions in pristine samples R-124, the pairing interaction V and the renormalized Coulomb interaction U^*; superconductivity arises when $V>U^*$. Here we are considering s-wave superconductivity. In fact, p- or d-wave superconductivity may arise even when $U^*>V$. With the incorporation of Pr, a third interaction emerges, which is the exchange interaction as mentioned above.

We consider all three interactions in terms of the structural parameters of disordered systems through the dielectric function in the region encompassed by the nearest CuO_2 layers. If ϵ_i denotes the dielectric constant of the $i = 1\text{-}2\text{-}3$ or 1-2-4 system, we may write

$$V = V_0/\epsilon_i, \quad U^* = U_0^*/\epsilon_i, \quad J_{SF} = J_{0SF}/\epsilon_i \ . \tag{8}$$

Here V_0, U_0^*, and J_{0SF} are the bare interactions which are assumed to be same for both R-123 and R-124 systems. The effects of the different crystal structure appear through the respective dielectric constants ϵ_{123} and ϵ_{124}. Two distinct regions can be identified in the structures of R-123 and R-124. Region 1 is enclosed by two nearest-neighbor CuO_2 planes and involves Y, Eu or Pr ions. The CuO_2-CuO_2 interplanar distance in region 1 is $a_{1i} \sim 4$ Å. Region 2 is bounded by two neighboring CuO_2 planes and involves the Ba ions. The CuO_2-CuO_2 distance here (region 2) is $a_{2i} \sim 8$ Å for the $i = 1\text{-}2\text{-}3$ and ~ 10 Å for the $i = 1\text{-}2\text{-}4$ systems (Fig. 6). We now consider the hopping of the holes along and perpendicular to the CuO_2 planes. $t_\parallel$ and $t_{\perp i}$ denote the corresponding hopping integrals. For simplicity we assume $t_\parallel$ to be same for both the $i = 1\text{-}2\text{-}3$ and $i = 1\text{-}2\text{-}4$ systems and that the perpendicular hopping occurs between the regions of type 1, so that the hole may be considered to be localized in a range a_{1i} (along the perpendicular direction). We further assume that hopping along the z direction is due to Anderson decay so that

$$t_{\perp i} \propto e^{-a_{2i}/a_{1i}} \ . \tag{9}$$

To see the connection between $t_{\perp i}$ and ϵ_i we consider the variation of the effective carrier concentration in the regions of type 1 with variation of $t_{\perp i}$. For $t_{\perp i} = 0$, the holes will remain localized in the regions of type 1. For $t_{\perp i} > 0$ holes will spend some time in regions of type 2 so that the effective concentration would decrease in regions of type 1. This would mean the effective (time-averaged) hole concentration would be higher in regions of type 1 for lower $t_{\perp i}$. Accordingly, the time-averaged dielectric constant as experienced by the holes in region 1 would also increase with decreasing $t_{\perp i}$. We may thus write

$$\epsilon_i = (t_\parallel / t_\perp)^p \ . \tag{10}$$

Here $p\,(>0)$ is a phenomenological constant.

The effective interaction $V\text{-}U^*$ in such a situation would decrease with increasing ϵ_i. From Eqs. (9) and (10) we find $\epsilon_{124} > \epsilon_{123}$, making $(V\text{-}U^*)_{124} < (V\text{-}U^*)_{123}$. This would lead to $T_c(124) < T_c(123)$, which indeed is the observed result for both the systems under consideration.

Turning to the effect of disorder, we see from Eqs. (8)–(10) that $J_{SF,124} < J_{SF,123}$. This means that the disorder will be less effective in RPr-124 than in RPr-123, leading to a higher x_{cr} value in the former case. In fact, assuming the density of states to be the same for 1-2-3 and 1-2-4 systems, we obtain

$$\frac{x_{cr,124}}{x_{cr,123}} = \left[\frac{t_{\perp,123}}{t_{\perp,124}} \right]^{p/2} = \exp\left[\frac{p\,(a_{2,124} - a_{2,123})}{2a_{1i}} \right] . \tag{11}$$

A value of $p \sim 0.45$ leads to $x_{cr,124} \sim 0.80$ for YPr-124 and $x_{cr,124} = 0.54$ for the EuPr-124 system. These values are quite close to the observed values. Realistic values of the density of states would only lead to a slightly different p parameter.

The way we have introduced the parameter p requires $p > 0$ only. There is no way to calculate the value of p; $p = 0.45$ is taken to obtain the observed values of x_{cr}. But if we note that

$$\frac{x_{cr,124}}{x_{cr,123}} \approx \frac{N_{CuO,124}}{N_{CuO,123}} \ , \tag{12}$$

where $N_{CuO,i}$ ($i = 1\text{-}2\text{-}4$, 1-2-3) is the total number of Cu and O atoms per (Y,Eu)Pr atom in the system ($N_{CuO,123} = 10$ and $N_{CuO,124} = 12$), we may attach some significance to the $p = 0.45$ value. Equation (9) means that Cu and O atoms of the CuO_2 planes and the CuO chains share the Pr atoms in terms of the pair-breaking effect. This provides a physical significance to the parameter p in the sense that the effect of Pr concentration absorbed by the effective pairing interaction $(V\text{-}U^*)$ is proportional to the total number of Cu and O atoms per $R_{1-x}Pr_x$ atom. More precisely, p indicates the departure from the linear dependence of the Pr concentration on N_{CuO}. Whether such an interpretation of p is universal or is particular to the systems considered here may be tested by considering some other systems, say, $Y_2Ba_4Cu_7O_{14}$ with Pr, where $x_{cr} = 0.7$ and $x_{cr} = 0.48$ are expected for the Y- and the Eu-based systems, respectively.

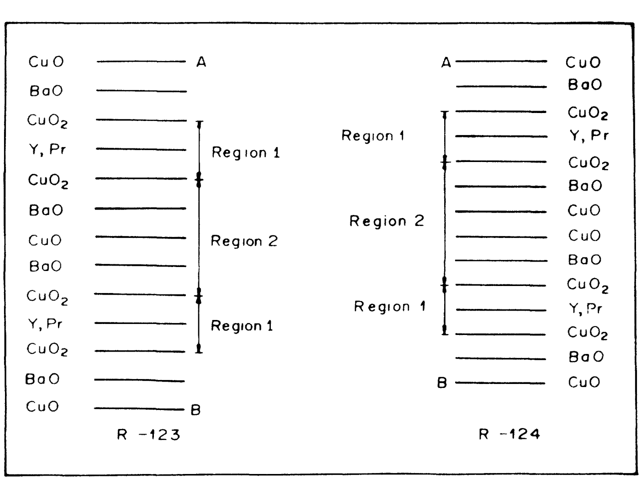

FIG. 6. Different layers of R-123 and R-124 structures depicting the regions 1 and 2. The separation of the layers marked *A* and *B* is 23.34 Å for Y-123 and 27.16 Å for Y-124.

V. CONCLUSIONS

A systematic study of the superconducting properties of the $(Y,Eu)_{1-x}Pr_xBa_2Cu_4O_8$ system is reported here. While the relative behavior of dT_c/dx in these systems is similar to that in $(Y,Eu)_{1-x}Pr_xBa_2Cu_3O_7$ systems, their T_c variation with x is more complicated. In particular, we find the Abrikosov-Gorkov pair-breaking theory to be inadequate for explaining the T_c behavior in the present case. A phenomenological modification of the AG theory on the basis of the observed resistivity leads to a better understanding of the situation in the YPr-124 system. Pr ions are found to induce an indirect coupling between the (Y,Eu) states and the Cu $3d$–O $2p$ state. This fact provides a reasonable explanation of the different T_c depression in the YPr-124 and EuPr-124 systems. The difference in the effects of Pr incorporation in the R-123 and R-124 families is expected to arise from the different distances between the neighboring CuO_2-(Y,Eu/Pr)-CuO_2 regions.

Note added in proof. After submitting this paper, we became aware of the work of Z. Guo, N. Yamada, K. I. Gondaira, T. Iri, and K. Kohn [Physica C **220**, 41 (1994)] on the T_c variation in the $Y_{1-x}Pr_xBa_2Cu_4O_8$ compound. The qualitative behavior of the variation of T_c with x observed by us agrees with that of Guo *et al.* with a small quantitative difference for low x. The critical Pr concentration x_{cr} is also almost the same in both studies.

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