Pressure effect on T_c for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$

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The pressure effects on the superconducting transition temperature (dT_c/dP) have been systematically measured for a series of $(Yb_{1-x}Pr_x)Ba_2Cu_3O_{7-\delta}$ with x ranging from 0 to 0.3. It has been found that $dT_c/dP<0$ for $x=0$, 0.2, and 0.3, and $dT_c/dP>0$ for $x=0.05$, 0.075, 0.1, and 0.15. We have unexpectedly observed a pressure-induced increase in the normal state resistivity for $x=0.2$ and 0.3, suggesting a decrease of the carrier concentration under pressure. Making use of the concept of pressure-induced charge transfer incorporated in the model of hole depletion by Pr doping, we have proposed a phenomenological model, which has successfully explained the x dependences of dT_c/dP and T_c.

I. INTRODUCTION

After the discoveries that $RBa_2Cu_3O_7$ (R-123) with[1] $R=$Y or rare earth element[2] are all 90 K superconductors except $R=$Pr, Ce, and Tb, many studies have been made to find why Pr is different from other rare earth in this system. As a collective effort on probing the mechanism of nonsuperconductivity in Pr-123,[3] it becomes clear that the hybridization of Pr $4f$ and O $2p$ orbits plays an essential role. However, the nature of this hybridization effect on the superconductivity is still not clear. In order to understand this nature, the dependence of the superconducting transition temperature T_c on Pr doping in $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ has been extensively studied.[4–8] The models for explaining this T_c-x relation include hole filling,[5] pair breaking,[5,6] hole localization,[7] and percolation.[8] Among them, the model of hole filling is directly related to the number of carriers (n) which is known to be a key parameter determining T_c. The results of the Hall measurement[9] and the Ca-doping experiment[5] on $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ showed that the presence of Pr was accompanied by a decrease of n, in consistence with the model of hole filling. On the other hand, many measurements[7,10,11] showed that the valence of the Pr is $+3$ and it was thus argued that a Pr ion could not possibly provide an electron to fill the hole in the CuO_2 plane. In a model of Fehrenbacher and Rice (FR),[12] it was suggested that the hole localizes at the Pr site and the extra valence in addition to $+3$ for the Pr ion may not be detectable by some optical measurements. This model resolves the controversy on the different valence values of the Pr ion obtained from different measurements. But, it still cannot explain the R dependence of T_c suppression in $(R_{1-x}Pr_x)Ba_2Cu_3O_7$.[13] Liechtenstein and Mazin (LM),[14] therefore, have modified the work of FR, and proposed that the L orbit in Pr-123 forms a hole-depleting band, which crosses the Fermi level and grabs holes from the CuO $pd\sigma$ band. On doping Y (or other R) in Pr-123, the position of this hole-depleting band shifts with R, resulting in the R dependence of T_c suppression.

According to the work[15] on the electronic structure of Gd-123 and Nd-123, the substitution of Gd with Nd reduces the energy difference between $4f$ and conduction electrons, and, hence, strengthens the hybridization effect of $4f$ and conduction electrons. Our previous high-pressure study[16] on R-123 has further revealed that the R substitution induces a charge redistribution giving rise to a T_c variation in the series of R-123. We have therefore decided to investigate the combined effort of chemical pressure (R-size effect) and physical pressure for $(R_{1-x}Pr_x)Ba_2Cu_3O_7$, hoping to shed some light on the mechanism of the nonsuperconductivity in Pr-123.

It has been demonstrated that high pressure can influence T_c greatly.[17] According to the proposed model of pressure-induced charge transfer (PICT), there are two contributions to the pressure effect of T_c (dT_c/dP). One is the pressure effect on the maximum T_c and the other is the pressure effect on the carrier concentration. In addition, it was further suggested[17] that dT_c/dP is positive for an underdoped sample (when the carrier concentration of the sample is lower than the optimal value), and negative for an overdoped sample (when the carrier concentration of the sample is higher than the optimal value). However, the pressure work of $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ (Ref. 18) and $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$ (Refs. 19 and 20) have shown that dT_c/dP is negative for the samples with $x>0.3$, which require a modification of the PICT model since the samples are not overdoped but with a negative dT_c/dP.

In this work, we have applied physical pressure on a series of $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ to measure the pressure dependence of T_c depression at various Pr-doping levels (x). The

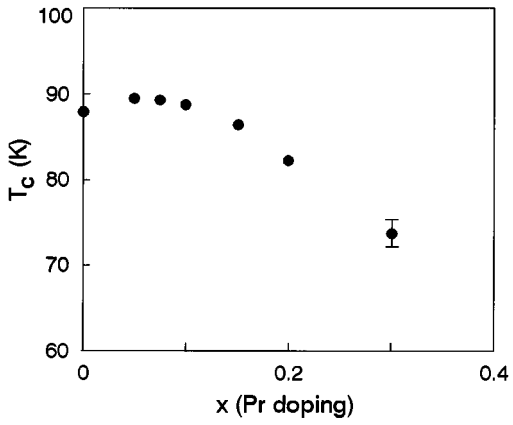

FIG. 1. T_c vs x (Pr-doping level) for various $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$. The solid circles represent the measured T_c values with the vertical bars showing the transition widths. The transition width is less than or equal to the size of solid circles for $x<0.15$.

reason for choosing Yb system is that the ionic radius of Yb is the second smallest while that of Pr is the largest in the rare-earth series. The larger difference in their ionic sizes gives rise to a plateau on the T_c-x curve in $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ at small x.[6] The origin of this plateau for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ will also be explained in this paper. Our experimental data have shown that pressure enhances T_c for samples with low x ($0<x<0.2$) but suppresses T_c for those with higher x ($x=0.2$ and 0.3). These results will be discussed based on a modified model of PICT with a concept of pressure-enhanced hole depletion.

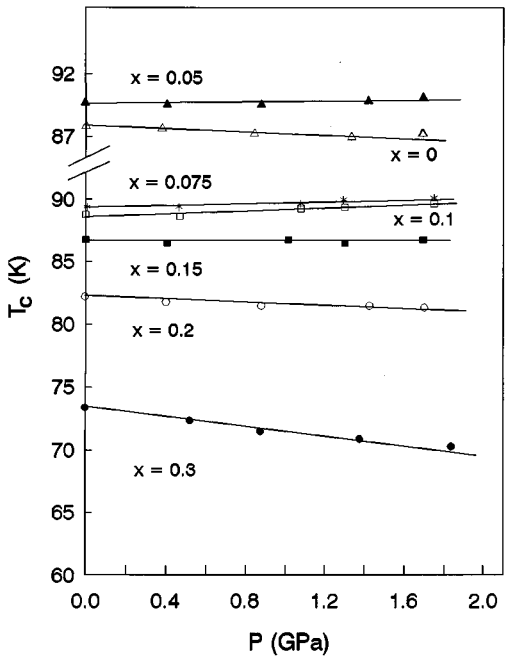

FIG. 2. T_c vs P for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ with x increasing from 0 to 0.3 (note: there is a break in the y axis for the values of T_c between $x=0$ and $x=0.075$).

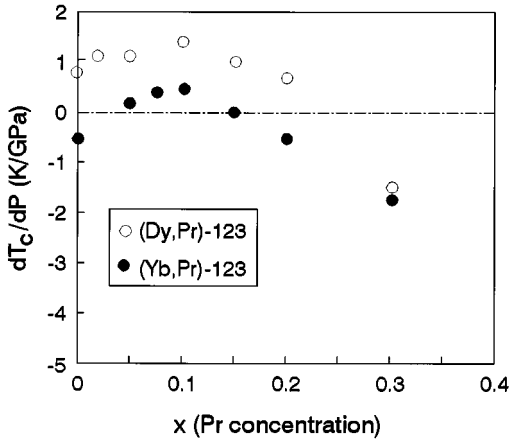

FIG. 3. dT_c/dP vs x for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ (the solid circles) and $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$ (the open circles). Data for $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$ are taken from Ref. 19.

II. EXPERIMENTS

Polycrystalline $(Yb_{1-x}Pr_x)Ba_2Cu_3O_{7-\delta}$ with $x=0.0$, 0.05, 0.075, 0.1, 0.15, 0.2, and 0.3 were prepared by the standard solid-state reaction technique. All samples were post annealed in flowing oxygen at 400 °C for 8 h. The oxygen contents of these samples were determined to be 6.97 ± 0.03 by using the idometric titration method. Structural characterization was carried out by using a Rigaku DMAX/BIII powder diffractometer to confirm the phase purity of the samples, and the dc magnetization at field cooling was measured to check their homogeneity. The sample was individually sealed in a Teflon cell with 3M Florinert fluid as a pressure medium and the cell was contained in a Be-Cu high pressure clamp[21] to generate hydrostatic pressure, P, up to 1.8 GPa. The four-point method was employed for resistivity measurements by using the standard inductance bridge. T_c was defined as the temperature at which the electrical resistivity drops to 50% of its extrapolated normal-state value. And, the transition width (ΔT_c) was defined as the temperature difference between 90% and 10% of the extrapolated normal-state resistivity values. It was found previously that the pressure effect of T_c depends critically on the sample quality, and the data are reliable only when the transitions are sharp. In this work, ΔT_c is less than 1.5 K for $x\leqslant0.2$, and 3.2 K for $x=0.3$.

III. RESULTS AND DISCUSSION

Figure 1 shows our data of T_c vs x for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$. The vertical bar represents the transition widths. For $x\leqslant0.2$, the transition widths are less than or equal to the size of the solid circles. T_c shows a nonlinear and nonmonotonous behavior for samples for $x<0.2$; while it increases slightly with x from 0 to $\sim$0.05, it, however, decreases from $\sim$0.05 to 0.3. This T_c-x relation is consistent with the previous results of Xu and Guan.[22,23]

T_c vs P for several $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ samples are shown in Fig. 2. T_c increases linearly with increasing pressure for $x=0.05$, 0.075, and 0.1, but decreases linearly for $x=0$, 0.2, and 0.3. Within our experimental accuracy, the crossover from a positive to a negative slope takes place at $x=0.15$, at which T_c is nearly constant with pressure up to

1.8 GPa. According to the least-square fit (see solid lines in Fig. 2), the slope of T_c vs P for $x=0$ is negative, then becomes positive and increases with increasing x until x reaches 0.075. From $x=0.075$ to 0.10 the slope is still positive but decreases with increasing x, and finally becomes negative at $x=0.2$ and 0.3. Similar results have been obtained for $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ (Ref. 18) and $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$,[19] but, in these cases, the values of dT_c/dP become negative at higher values of x.

In Fig. 3, we have plotted dT_c/dP for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ as a function of x, and, for comparison, we have included the data for $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$.[19] From these two sets of data, we have noticed three interesting features. First, for undoped samples ($x=0$), dT_c/dP is positive for Dy-123 but negative for Yb-123. Secondly, the nonlinear behavior of dT_c/dP vs x for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ is similar to that for $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$. Thirdly, the value of dT_c/dP becomes negative at large x for both systems. The opposite sign of dT_c/dP for Yb and Dy-123 has been explained in Ref. 14 by the nature of doping: overdoped for Yb-123 and underdoped for Dy-123, arising from the charge redistribution between the charge reservoir and the CuO_2 plane. For the second and the third features in Fig. 3, the similarity in these two dT_c/dP vs x curves suggests that there is a common mechanism, closely related to the mechanism of T_c depression by Pr doping, governs the pressure effect on T_c in $(R_{1-x}Pr_x)Ba_2Cu_3O_7$.

The nonlinear behavior of the T_c-x relation for small x (see Fig. 1) has also been observed in some small-R systems such as $(Er_{1-x}Pr_x)Ba_2Cu_3O_7$ or $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$, and has been attributed to a nonmagnetic-magnetic transition by Xu and Guan.[22] This model assumed that there was a region where the $4f$ electrons of Pr are heavily hybridized with the conduction band such that Pr loses its magnetic moment (as well as the pair-breaking strength), hence, T_c is unaffected by Pr doping in this region [the small x region in $(R_{1-x}Pr_x)Ba_2Cu_3O_7$ with smaller R]. When x crossed the proposed transition, their magnetic moment (and the related pair-breaking strength) recovered and T_c was suppressed linearly with Pr doping. This model can be tested in the present pressure study because the magnetic interaction is sensitive to the interatomic distance, as demonstrated in some previous reports on the conventional superconductors.[24] According to the model of Xu and Guan,[22] the change in the slope of T_c-x curve at $x=0.15$ for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ should be regarded as a transition from nonmagnetic to magnetic state. In the case of $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$, T_c-x curve, is, however, linear at $x=0.15$, and, hence, based on this model, the magnetic transition does not occur. Therefore, one should expect the dT_c/dP-x curve in $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ to be very different from that of $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$. Our dT_c/dP-x data, nonetheless, show a similar behavior for these two systems (see Fig. 3), suggesting that the model of Xu and Guan[22] is inadequate to explain these pressure data. To further confirm the absence of this nonmagnetic-to-magnetic transition in $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$, we have also measured the magnetic moment of the Pr ion for $(Y_{0.98}Pr_{0.02})Ba_2Cu_3O_7$ by using a superconducting quantum interference device magnetometer. Pr ion is expected to have zero magnetic moment if it is in the nonmagnetic region. A magnetic moment of 2.5 μ_B has been, however, obtained, which is in agreement with the values obtained for various $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ by other groups.[6,25] Our data implies that the model of the absence of the magnetic moment in Pr at small x is not likely to be the mechanism governing the observed plateau in the T_c-x relation for $(R_{1-x}Pr_x)Ba_2Cu_3O_7$ with smaller R.

A hole-filling/pair-breaking model has been proposed to interpret the x dependence of T_c and dT_c/dP in the $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ system by Maple *et al.*[18] In this model, the behavior of T_c-x at ambient pressure is interpreted as the outcome of the competition between a parabolic carrier-density term and a linear pair-breaking effect:

$$T_c = T_{c0} - A(\alpha - \beta x)^2 - Bx, \tag{1}$$

where A is a constant, B the pair-breaking parameter, and $(\alpha-\beta x)$ the number of effective carriers and it was assumed that only T_{c0}, β, and B changed with pressure. Then,

$$dT_c/dP = dT_{c0}/dP + (2A\alpha d\beta/dP - dB/dP)x - (2A\beta d\beta/dP)x^2. \tag{2}$$

However, it has been shown in Ref. 18 that owing to the large uncertainty on dB/dP, Eq. (2) could not be used to provide information on the pressure dependence of the magnetic interaction. In other words, although Eq. (1) describes the T_c-x relation of $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ rather well, Eq. (2), which is derived from Eq. (1), cannot satisfactorily explain the experimental data of dT_c/dP vs x.

In Fig. 3, our data show that dT_c/dP is negative for $x=0$, 0.2, and 0.3. For $x=0$ (pure Yb-123), negative dT_c/dP can be understood because it is in the overdoped state, in which an increase in n by pressure causes T_c decrease as seen in the previous report for (Y,La,Ca)-123.[17] The negative dT_c/dP values for $x=0.2$ and 0.3 are somehow unusual, since these two samples have low T_c and are not overdoped. According to the fitting for the pressure data of $(Y_{1-x}Pr_x)Ba_2Cu_3O_7$ by using Eq. (2),[18] $d\beta/dP$ is positive and dB/dP is negative, indicating that the negative dT_c/dP results from the quadratic term of x. In other words, the major contribution to the negative dT_c/dP is the increase of β with P (note: the increase in β means the decrease in the hole concentration). This observation is in accord with our results of the pressure dependence of the resistivity behavior. We have found that the normal state resistivity decreases under pressure for $x<0.2$, but increases under pressure for $x=0.2$ and 0.3. Figures 4 and 5 display the temperature dependence for resistivity ρ under various pressures for $x=0$ and 0.3, respectively. Figure 4 shows a decrease of the normal state ρ and a decrease in T_c under pressure, while in Fig. 5, the normal state ρ increases with a decrease in T_c under pressure. The value of $d\ln\rho/dP$ has been obtained to be $-10\pm2\%$/GPa for samples with $x<0.15$, which is consistent with those of high temperature superconductors (HTS). This value of $d\ln\rho/dP$ found in most HTS has been attributed to the pressure-induced charge transfer from the charge reservoir to the CuO_2 plane.[26] Therefore, the observed positive value of $d\ln\rho/dP$ in the normal state for $x=0.2$ and 0.3 are rather unusual in HTS and cannot be simply explained by the pair breaking effect, which occurs only in the superconducting state. If the model of pressure-induced charge transfer (PICT) is valid, however, positive $d\ln\rho/dP$ can be under-

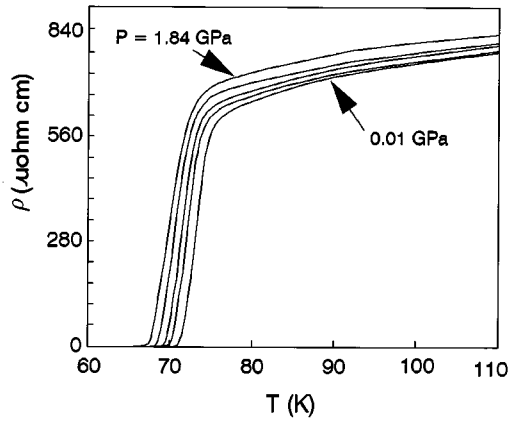

FIG. 4. Temperature dependence of ρ for pure Yb-123 at various pressures.

stood in terms of a decrease in n induced by pressure. However, for samples with large x, the ρ-T behavior may change from metalliclike to insulatinglike,[18–20] which could be due to a pressure-induced hole localization.[20]

The model of FR (Ref. 12) has shown that the mixed state of Pr^{+3} and Pr^{+4} plays an essential role in determining the electronic state of the CuO_2 plane. When the hole state in Pr-O_7 is more stable than in the Cu-O planar singlet, more holes are bound to Pr sites. More precisely, holes will transfer from the Cu-O $pd\sigma$ band into the FR state which is a mixture of $4f^1$ and $4f^2\underline{L}$ configurations with $\underline{L}$ being a ligand hole in the O $2p$ orbit. In this model, there exist two discrete hole states: $n_p=0$ or $n_p=(1-n_c)/2$ where n_p is the number of holes per CuO_2 plane and n_c the number of holes per CuO chain. Nonetheless, with only two discrete stable solutions for the hole states, this model cannot be used to explain the observed continuous suppression of T_c by increasing Pr doping. Therefore, Liechtenstein and Mazin[14] modified the work of FR by assuming an additional dispersive band crossing the Fermi level, which can grab holes from the Cu-O $pd\sigma$ band (namely, the fractional holes can be transferred to the FR state). The result of the calculation in the density of states[14] refers that the number of holes transferred to the FR state is x and R dependent.

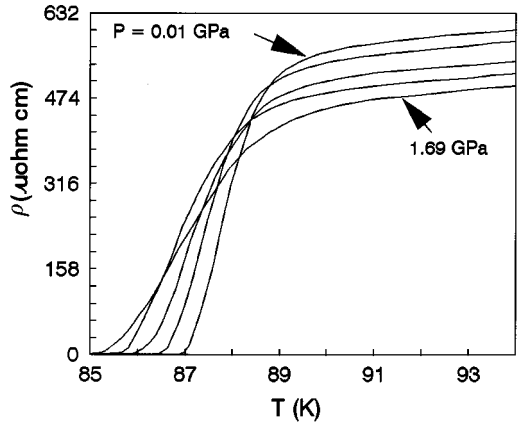

FIG. 5. Temperature dependence of ρ for $(Yb_{0.7}Pr_{0.3})Ba_2Cu_3O_7$ at various pressures.

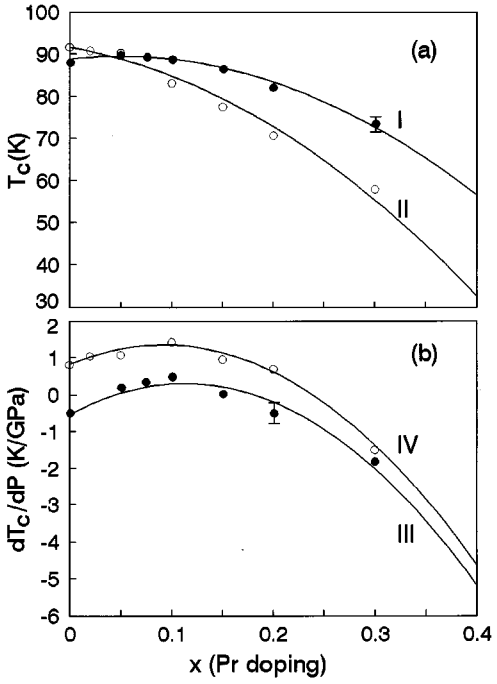

FIG. 6. (a) Solid curves are the theoretical fittings of T_c vs x for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ (curve I) and $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$ (curve II); (b) solid curves are the theoretical fittings of dT_c/dP vs x for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ (curve III) and $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$ (curve IV). In both figures, the vertical line represents the experimental error bar.

Based on the argument of LM,[14] the observed negative dT_c/dP accompanied by positive $d\ \ln\rho/dP$ can be explained in terms of the pressure-enhanced hole depletion from the CuO_2 $pd\sigma$ band, resulting in a pressure-induced T_c suppression. However, the positive dT_c/dP values for $0<x<0.15$ indicate the presence of another pressure effect on T_c, which is believed to be the effect of PICT. Consequently, in order to explain our data of T_c-x and dT_c/dP-x, we have proposed a model based on the theory of LM along with the concept of PICT. The central theme of our model is that the pressure effect on T_c arises from two competitive components: one is the PICT from the CuO_2 chain to the CuO_2 plane, yielding an increase in n; the other is the pressure-enhanced hole transfer from the CuO_2 plane to the FR state, giving a decrease in n.

Including the hole-depletion effect of Pr doping in the parabolic function of n, T_c can be written as

$$T_c=T_{c0}-C(n-n_0-Dx)^2, \tag{3}$$

where C is a constant, n_0 the optimal n corresponding to the maximum T_c (T_{c0}), and D an effective factor related to the hole depletion by each Pr ion. Here, we assume D to be pressure dependent. Then,

$$\begin{aligned} dT_c/dP &= dT_{c0}/dP-2C(n-n_0-Dx)dn/dP \\ &\quad +2Cx(n-n_0-Dx)(dD/dP) \\ &= dT_{c0}/dP-2C(n-n_0-Dx) \\ &\quad \times(dn/dP-xdD/dP), \end{aligned} \tag{4}$$

in which dn/dP is the rate change of the number of carriers under pressure through charge transfer from the CuO chain to the CuO_2 plane, and xdD/dP is that from the CuO_2 plane to the FR state. We have used Eq. (3) to fit our experimental data of T_c vs x with T_{c0} and D fixed, allowing n_0-n and C to vary. T_{c0} is chosen to be 89.6 K because it is the maximum T_c in the T_c-x curve (see Fig. 1), and we have taken $D=0.3$ as suggested by LM.[14] As shown in Fig. 6(a), curve I is our best fit for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ with $C=3000$ and $n-n_0=0.015$. For comparison, we have included the data and the fitting (curve II) for $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$. For curve II, $C=3000$, $n_0-n=0.022$. From Fig. 6(a), we have found that curve II is more linear than curve I, in consistence with the experimental observations for those systems with large R.[6,22,23] Inserting the same values of n_0-n, C, and D as those used to obtain curves I and II, the data dT_c/dP vs x were fitted by using Eq. (4). We have employed $dn/dP=0.0065$ hole/GPa following Ref. 27, with dD/dP and dT_{c0}/dP to be the adjustable parameters. The physical meaning of dD/dP is the number of the pressure-enhanced hole depletion per Pr ion. Curve III is a fit with $dT_{c0}/dP=(0.05\pm0.03)$ K/GPa and $dD/dP=(0.037\pm0.007)$ GPa^{-1} for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$; curve IV is with $dT_{c0}/dP=(0.8\pm0.1)$ K/GPa and $dD/dP=(0.035\pm0.003)$ GPa^{-1} for $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$. As shown in Figs. 6(a) and 6(b), our fittings agree with the data within the experimental error. The good agreement between the experimental data and the theoretical curves suggests the applicability of our model.

IV. CONCLUSION

We have systematically measured T_c and dT_c/dP for the series of $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$. Our results have shown $dT_c/dP<0$ for $x=0$, 0.2, and 0.3 and >0 for $x=0.05$ to 0.15. The negative values of dT_c/dP for $x=0.2$ and 0.3 were accompanied by positive $d\ln\rho/dP$, suggesting a pressure-induced decrease in the carrier concentration. In order to explain our data, we have developed a phenomenological model by incorporating the concept of pressure-induced charge transfer into the model of LM. A good agreement has been obtained between the experimental data and the theoretical curves of T_c vs x and dT_c/dP vs x for $(Yb_{1-x}Pr_x)Ba_2Cu_3O_7$ and $(Dy_{1-x}Pr_x)Ba_2Cu_3O_7$. For this reason, our model can also be used to predict the R dependence of dT_c/dP vs x. Nevertheless, further testing of this model calls for more pressure work on other $(R_{1-x}Pr_x)Ba_2Cu_3O_7$.

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