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Effects of Pr ion on the superconducting properties in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ single crystals

Yi Zhuo,* Jae-Hyuk Choi, Mun-Seog Kim, Jin-Nam Park, Myoung-Kwang Bae, and Sung-Ik Lee
Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Republic of Korea

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$Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ single crystals with $x = 0.1$, 0.2, and 0.3 were grown by the flux method. The temperature dependence of the magnetization with the external magnetic field parallel to the c axis was measured with a superconducting quantum interference device magnetometer. In the thermodynamically reversible region, the magnetization was analyzed and the various thermodynamic parameters were obtained. The upper critical field slope dH_{c2}/dT near T_c, the zero temperature upper critical field $H_{c2}(0)$, and the superconducting transition temperature T_c decreased monotonically with increasing Pr concentration. Other parameters such as the Ginzburg-Landau parameter κ, the penetration depth $\lambda_{ab}(0)$, and the coherence length $\xi_{ab}(0)$ increased with Pr content. The effects of Pr ion on the superconducting properties are discussed. [S0163-1829(97)07834-X]

I. INTRODUCTION

It is well known that superconductivity is strongly suppressed by the substitution of Pr for Y in $YBa_2Cu_3O_{7-\delta}$ (YBCO).[1–5] The $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ system is isostructural with YBCO, but T_c decreases monotonically with increasing x. T_c reaches zero at $x \approx 0.56$.[6] This depression rate is strongly compared with that induced by the substitution of other rare-earth elements R (except Ce, Tb) for Y of YBCO, where T_c does not change. Several suggestions, such as magnetic pair breaking, hole localization, or hole filling, have been proposed to explain the suppression of T_c.[1,3,4,7–13,35,36] Related to the mechanism of T_c suppression, intense attention has been called to the issue of the valence state of Pr in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$, but considerable controversy still persists over it.[6,10,12–17]

In order to understand the suppression of T_c, systematic studies of the microscopic physical quantities are quite necessary. Until now, the effect of Pr on the thermodynamic parameters such as the Ginzburg-Landau parameter κ, the coherence length ξ, and the magnetic field penetration depth λ have not been studied in detail. These quantities reflect the intrinsic characteristics of the superconductor.

A single-crystal sample is the best candidate to study the microscopic mechanisms of superconductivity, although the small size of the sample often makes the experiment very delicate. In this paper, we have prepared $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ single crystals with $x = 0.1$, 0.2, and 0.3 (called YPr-0.1, YPr-0.2, and YPr-0.3, respectively), and measured the temperature dependence of magnetization in various magnetic fields applied along the c axis. We have analyzed the reversible magnetization using the Hao-Clem model and have obtained a series of superconducting parameters with different x.[18,19]

II. EXPERIMENTS

Single crystals were grown by the flux method.[21] First, the polycrystalline precursors of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ were prepared. The precursors were mixed with an excess of $BaCO_3$ and CuO powder. Finally, the crystals with typical dimensions of $1.5\times1.5\times0.05$ mm^3 were obtained. All of the crystals were annealed in oxygen at 400 °C for a week. The magnetic properties of the single crystals were measured with a superconducting quantum interference device (SQUID) magnetometer. The measurements in the zero-field-cooled and field-cooled conditions were repeated at various magnetic fields parallel to the c axis. To obtain the intrinsic $M(T)$, the normal-state background was appropriately subtracted from the observed magnetization values in the high-temperature region of 150 K $\leq T \leq$ 250 K.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of magnetization at $H = 10$ Oe for single crystals with $0.1 \leq x \leq 0.3$. The superconducting transition temperature T_c decreases with increasing x as expected. The transition widths of these single crystals are relatively narrow compared to those of the polycrystalline compound, which indicates the synthesis of high-quality crystals.

Figure 2 shows the temperature dependence of the reversible magnetization for magnetic fields parallel to the c axis. For the same external field, the diamagnetism below T_c decreases with increasing Pr concentration, which indicates the

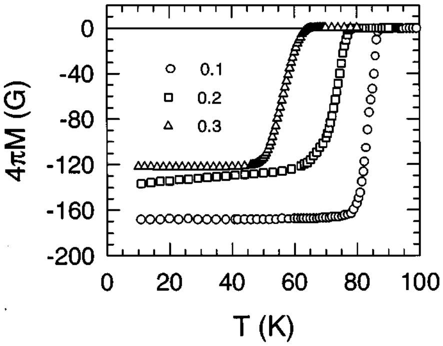

FIG. 1. Temperature dependence of the magnetization with an external magnetic field of $H = 10$ Oe (zero-field cooling, $H \| c$) for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ with $x = 0.1$, 0.2, and 0.3.

suppression of superconductivity from the Pr ion. In addition, the crossover of magnetization is observed at 1~2 K below T_c for all samples. This phenomenon, due to vortex fluctuation, was also observed in YBCO and Hg-based superconductor.[23–25] This is more obvious for highly anisotropic Bi- or Tl-based oxide superconductors.[22,26,27]

For the high-T_c superconductors, there exists a broad intermediate magnetic field region ($H_{c1} \leq H \leq H_{c2}$) in which most magnetic properties were investigated. In this region, the conventionally accepted London model is not valid because it ignores the core energy contribution to the magnetization.[28–30] Hao and Clem[19] take in account this energy into the total free energy and calculate a reversible magnetization in the entire field region between H_{c1} and H_{c2}.

In the Hao-Clem model, the dimensionless magnetization $-4\pi M' = -4\pi M/\sqrt{2}H_c(T)$ is expressed as[19]

$$-4\pi M' = \frac{1}{2}\frac{\partial}{\partial B}(F-2B^2)_{f_\infty,\xi_v}$$

$$= \frac{\kappa f_\infty^2 \xi_v^2}{2}\left[\frac{1-f_\infty^2}{2}\ln\left(\frac{2}{B\kappa\xi_v^2}+1\right) - \frac{1-f_\infty^2}{2+B\kappa\xi_v^2} + \frac{f_\infty^2}{(2+B\kappa\xi_v^2)^2}\right] + \frac{f_\infty^2(2+3B\kappa\xi_v^2)}{2\kappa(2+B\kappa\xi_v^2)^3}$$

$$+ \frac{f_\infty}{2\kappa\xi_v K_1(f_\infty\xi_v)}\left(K_0[\xi_v(f_\infty^2+2B\kappa)^{1/2}] - \frac{B\kappa\xi_v K_1[\xi_v(f_\infty^2+2B\kappa)^{1/2}]}{(f_\infty^2+2B\kappa)^{1/2}}\right), \tag{1}$$

where $K_n(x)$ is a modified Bessel function of nth order, and ξ_v and f_∞ are two variational parameters representing the effective core radius of a vortex and the depression of the order parameter due to overlapping of vortices, respectively. The two variational parameters f_∞ and ξ_v are approximately written as

$$f_\infty^2 = 1 - \left[\frac{B}{\kappa}\right]^4, \tag{2}$$

$$\left[\frac{\xi_v}{\xi_{v_0}}\right]^2 = \left[1 - 2\left(1-\frac{B}{\kappa}\right)^2\frac{B}{\kappa}\right]\left[1+\left(\frac{B}{\kappa}\right)^4\right], \tag{3}$$

for the cases of $\kappa > 10$ with $\kappa\xi_{v_0} \cong \sqrt{2}$.

Equation (1) is used to analyze the experimental data. First of all, a set of data $[-4\pi M_i, H_i]$ $(i=1,2,\ldots)$ is chosen at each fixed temperature from the reversible region. The detailed description of this procedure has been given in Refs. 8,12. The Ginzburg-Landau parameter $\kappa(T)$ and thermodynamic critical fields can be evaluated from the theoretical analysis. Figure 3 shows the temperature dependence of the Ginzburg-Landau parameter $\kappa(T)$. The $\kappa(T)$ decreases weakly with increasing temperature for $x = 0.3$ in the temperature range 52 K $\leq T \leq$ 55 K in addition to the general increase with temperature for all three samples. The $\kappa(T)$ of YPr-0.3 in the temperature range 52 K $\leq T \leq$ 55 K shows the typical behavior of type-II superconductors,[31] while the general increase of $\kappa(T)$ with temperature is due to the influence of the positional fluctuation of the vortices which the Hao-Clem model does not consider[19,20].

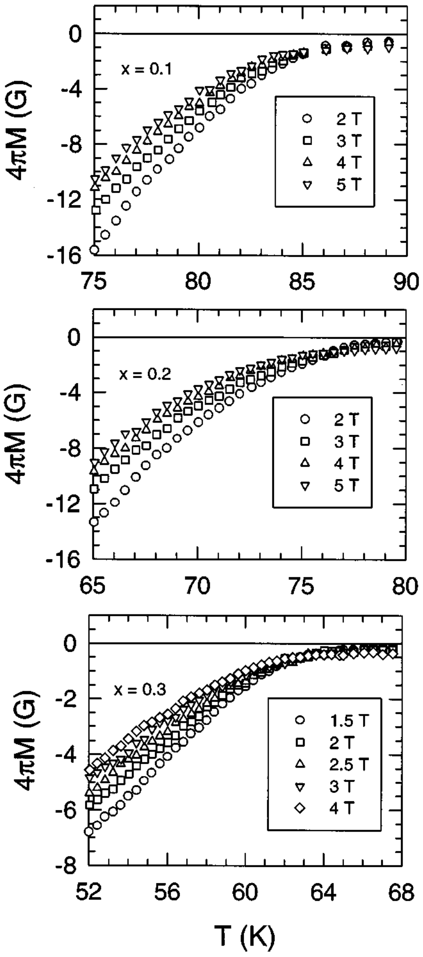

FIG. 2. Temperature dependence of the reversible magnetization $4\pi M(T)$ with applied field parallel to the c axis for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$.

For YPr-0.1, the $\kappa(T)$ value is nearly constant at the temperature region of 75 K $\leq T \leq$ 80 K. The average value of κ(T), κ_{av}, is 79 in this temperature range. In the same way, κ_{av} is 88 at the temperature region 66 K $\leq T \leq$ 72 K for YPr-0.2 and is 101 at the temperature region 52 K $\leq T \leq$ 58 K for YPr-0.3. The Ginzburg-Landau parameter κ increases with x.

The thermodynamic critical field $H_c(T)$ can be derived directly from the above analysis. Figure 4 shows $-4\pi M' = -4\pi M/\sqrt{2}H_c(T)$ versus $H' = H/\sqrt{2}H_c(T)$ of the experimental data and theoretical curve. The experimental data $[-4\pi M_i, H_i]$ at various temperatures collapses onto a universal curve with single-valued κ. Rather good agreement is obtained between the experimental data and theoretical predictions. Insets in Fig. 4 show the temperature dependence of $H_c(T)$ compared with the BCS result[32]

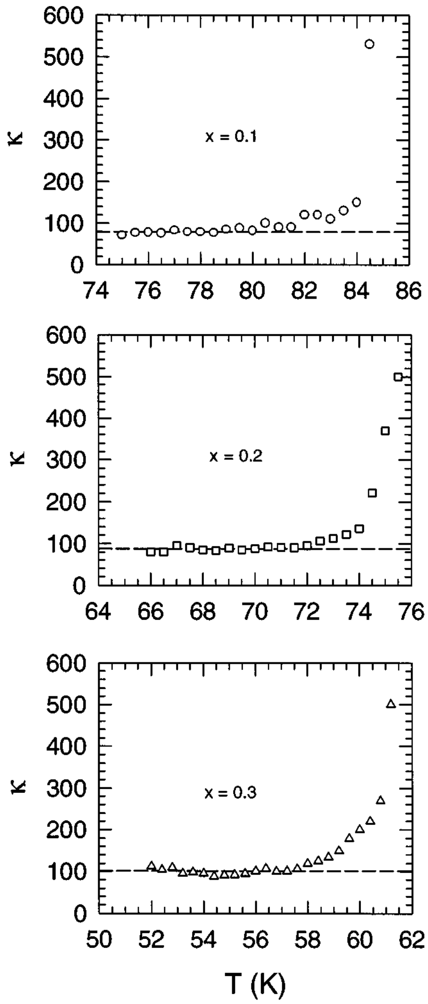

FIG. 3. Temperature dependence of the Ginzburg-Landau parameter $\kappa(T)$ from theoretical analysis for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$.

$$\frac{H_c(T)}{H_c(0)}=1.7367\left[1-\frac{T}{T_c}\right]\left[1-0.2730\left(1-\frac{T}{T_c}\right)-0.0949\left(1-\frac{T}{T_c}\right)^2\right]. \qquad (4)$$

From this analysis, $H_c(0)$ = 0.78 T with T_c = 86.6 K is obtained for YPr-0.1. For YPr-0.2, and YPr-0.3, $H_c(0)$ is 0.57 T with T_c = 77.8 K and 0.31 T with T_c = 64.1 K, respectively. These T_c values are comparable with the superconducting onset temperature determined from the low field measurements.

According to the relation $H_{c2}(T)=\sqrt{2}\kappa H_c(T)$, we find that $(dH_{c2}/dT)_{T_c}$ is -1.64 T/K, -1.48 T/K, and -1.09 T/K for YPr-0.1, YPr-0.2, and YPr-0.3, respectively. The $H_{c2}(0)$ is estimated from the equation[31]

$$H_{c2}(0)=0.5758\left[\frac{\kappa_1(0)}{\kappa}\right]T_c\left|\frac{dH_{c_2}}{dT}\right|_{T_c}, \qquad (5)$$

where $\kappa_1(0)/\kappa$ equals 1.20 in the dirty limit[31] and 1.26 in the clean limit.[33] For YPr-0.1, $H_{c2}(0)$ is 98 T in the dirty limit and 103 T in the clean limit. $H_{c2}(0)$ is 80 T (84 T), 48 T (51 T) in the dirty (clean) limit for YPr-0.2 and YPr-0.3, respectively. Compared with the case of pure YBCO, these parameters $[(dH_{c2}/dT)_{T_c}$, $H_c(0)$, $H_{c2}(0)$, $T_c]$ decrease monotonically with increasing Pr concentration (Table I). The systematic changes of these superconducting parameters are consistent with the results obtained from resistivity measurements,[34] which reflect the suppression of superconductivity.

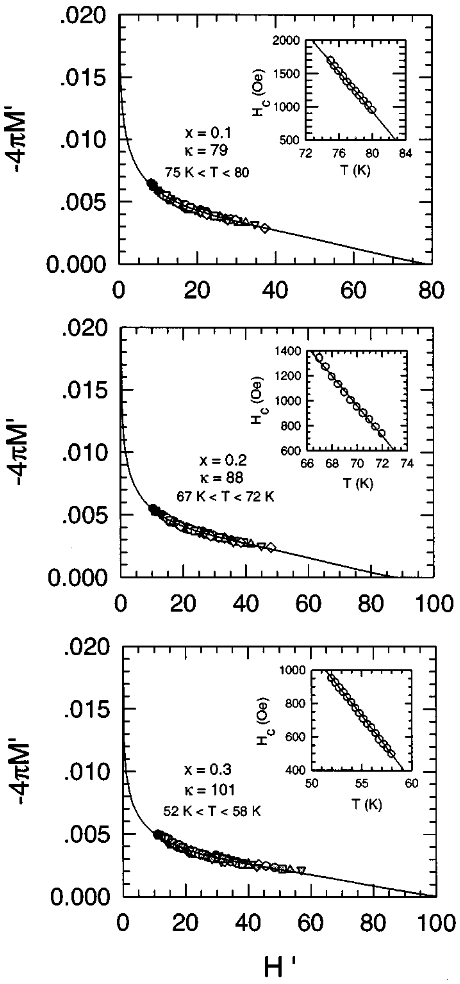

FIG. 4. Magnetization vs applied field scaled by $\sqrt{2}H_c(T)$ for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$. Solid line represents theoretical curve. Inset: Temperature dependence of thermodynamic critical field $H_c(T)$ obtained from the Hao-Clem model. Solid lines represent the BCS temperature dependence of $H_c(T)$.

The zero-temperature coherence length $\xi_{ab}(0)$ can be estimated from the expression $H_{c2}(0)=\phi_0/2\pi\xi_{ab}^2(0)$ without considering the anisotropy in the ab-plane.[29] The values $\xi_{ab}(0)$ = 18.3 Å (17.9 Å), 20.3 Å (19.8 Å), and 26.1 Å (25.5 Å) in the dirty (clean) limit are obtained for YPr-0.1, YPr-0.2, and YPr-0.3, respectively.

The magnetic penetration depth $\lambda_{ab}(T)$ is calculated through the relation $\sqrt{2}H_c(T)=\kappa\phi_0/2\pi\lambda_{ab}^2(T)$.[29] Figure 5 shows the penetration depth $\lambda_{ab}(T)$ and the theoretical curves of BCS dirty and clean limits. From this analysis, we obtain the zero-temperature magnetic penetration depth $\lambda_{ab}(0)$ = 1870 Å (1650 Å), 2300 Å (2040 Å), and 3340

TABLE I. Thermodynamic parameters of YBCO, YPr-0.1, YPr-0.2, and YPr-0.3 obtained from the Hao-Clem model analysis.

	YBCO	YPr-0.1	YPr-0.2	YPr-0.3
κ	57 (Ref. 10)	79	88	101
T_c (K)	94.1 (Ref. 10)	86.6	77.8	64.1
$H_c(0)$ (T)	1.10 (Ref. 10)	0.78	0.57	0.31
$(dH_{c2}/dT)_{T_c}$ (T/K)	−1.65 (Ref. 10)	−1.64	−1.48	−1.09
$H_{c2}(0)$ (T)	112 (Ref. 10)	103[a]	84[a]	51[a]
		98[b]	80[b]	48[b]
$\xi_{ab}(0)$ (Å)	17.2[a] (Ref. 10)	17.9[a]	19.8[a]	25.5[a]
	17.6[b] (Ref. 10)	18.3[b]	20.3[b]	26.1[b]
$\lambda_{ab}(0)$ (Å)	1310 (Ref. 37)	1650[a]	2040[a]	2980[a]
		1870[b]	2300[b]	3340[b]

[a] Clean limit.

[b] Dirty limit.

Å (2980 Å) in the dirty (clean) limit for YPr-0.1, YPr-0.2, and YPr-0.3, respectively. The penetration depth $\lambda_{ab}(0)$ increases with the Pr concentration.

From the above results, we notice that all three parameters [κ, $\lambda_{ab}(0)$, and $\xi_{ab}(0)$] increase with the Pr content. The increment of $\lambda_{ab}(0)$ with x in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ is consistent with the results of the Seaman *et al.* muon-spin-relaxation measurements[35] but is contrary to that of the Peng *et al.* magnetization measurements.[3] In the Peng *et al.* magnetization measurements, the samples were polycrystalline and not aligned. For this reason, the derived penetration depth represented the angular-averaged value. In their analysis, $H_{c2}(T)$ was taken as the value where the measured magnetization deviated from the high-field straight line. These $H_{c2}(T)$ curves near T_c were used to estimate the various thermodynamic parameters. But, as Hao *et al.* pointed out,[19] the determination of $H_{c2}(T)$ from linear extrapolation of the M vs T curve is not appropriate in applied magnetic fields H far below $H_{c2}(T)$. So, the deduced $\lambda_{ab}(0)$ may deviate largely from the real value.

One way to explain the increase of penetration depth is by its relationship with the charge carrier density. It is known that the zero-temperature penetration depth is proportional to $(m_{ab}^*/n)^{1/2}$ for the clean superconductor, where m_{ab}^* is the effective mass of the Cooper pair in the ab plane, and n is the superconducting carrier density.[29] A decrease of n could directly result in the increase of the penetration depth if the m_{ab}^* is not sensitive to x. It was reported from the Hall-effect measurements that the n decreases with increasing Pr concentration.[14,36] In addition, the deduced results from the normal state magnetic susceptibility also show that the Pr valence is greater than +3.[6] The substitution of Pr with this valence for trivalent Y may bring extra electrons to fill holes in the CuO_2 planes, i.e., the decrease of n, which could cause the increase of the penetration and the depression of superconductivity. This is consistent with our result. Certainly, the change of the penetration depth alone is not a sole explanation of the superconductivity suppression, and more detailed studies are needed.

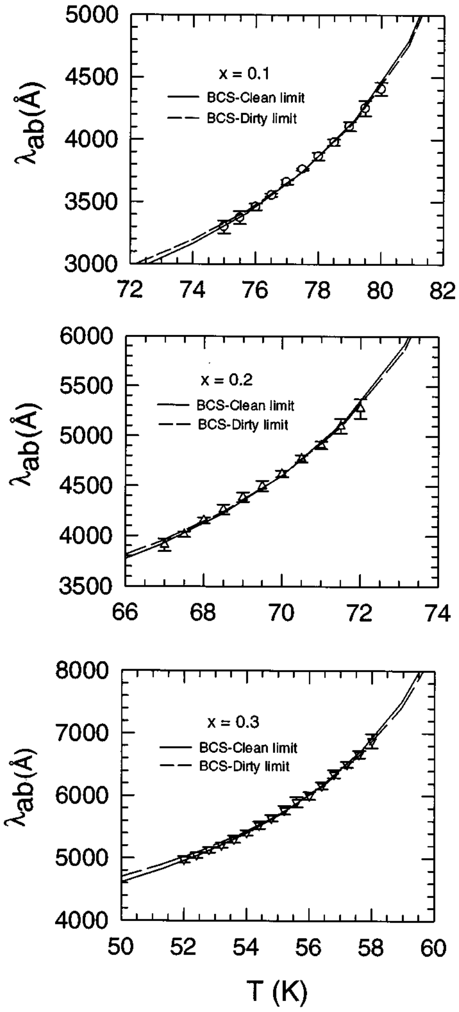

FIG. 5. Temperature dependence of the penetration depth $\lambda_{ab}(T)$ obtained from the theoretical fitting. Solid and dashed lines represent the BCS clean and dirty limits, respectively.

IV. CONCLUSION

We measured the high-field magnetization for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ (x = 0.1, 0.2, and 0.3) single crystals with applied fields along the c-axis. By applying the Hao-Clem model, we studied the effects of the Pr doping on the thermodynamic parameters. The obvious change of the parameters was observed. The results show that the upper critical field $H_{c2}(0)$ decreases with Pr concentration, while the parameters, such as κ, $\lambda_{ab}(0)$, and $\xi_{ab}(0)$, increase with Pr doping. Based on the change in $\lambda_{ab}(0)$, the mixed-valence state of Pr ion is most likely responsible for the suppression of superconductivity in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$.

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*Also at Department of Physics, Northeastern University, Shenyang 110006, P. R. China.

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