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STUDY OF OXYGEN ISOTOPE EFFECT IN Pr, Ca, AND Zn DOPED SUPERCONDUCTIVITY $YBa_2Cu_3O_{7-\delta}$,

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ABSTRACT

The oxygen isotope effect in Pr, Ca, and Zn doped superconductivity $YBa_2Cu_3O_{7-\delta}$ was investigated. Pr and Ca substitute predominantly at the Y site while Zn goes into the Cu plane site. The shift in critical temperature (ΔT_C) between samples oxygenated in ^{18}O and ^{16}O was obtained via dc resistance measurements, and low field dc magnetization and ac susceptibility measurements in a SQUID magnetometer. Confirmation of the substitution of the oxygen was a achieved with Raman and SIMS measurements. The Pr, Ca and Zn substituted change T_C of the superconductor in different ways. Increasing Pr concentrations lower T_C and increase ΔT_C with the isotope coefficient, α, approaching $1/2$. The additions of ca reduces the size of both ΔT_c and α. Both exhibit a small parabolic effect with increasing Ca substitution. An increase in Zn substitution. An increase in Zn substituted lowers T_C but ΔT_C remains nearly constant, or perhaps gets slightly smaller, with α increasing to approximately $1/3$.

KEYWORDS: Oxygen Isotope Effect in Pr, Ca, and Zn Doped Superconductivity $YBa_2Cu_3O_{7-\delta}$

INTRODUCTION

Calcium Substitution

Sample Preparation

The preparation method of the substitutions of 5% to 25% Ca, with a constant 20% Pr, into $Y_{0.8}Pr_{0.2}Ba_2Cu3O_{7-\delta}$ is different from the Pr substituted samples and thus a 0% Ca substituted sample is included as a comparison for the previous set of measurements. Mixed powders of $(Y_{0.8-Y}Pr_{0.2}Ca_Y)Ba_2Cu_3O_{7-\delta}$ for the concentrations of y = 0.05, 0.1,0.15, 0.2, and 0.25 were calcined in air at 905• C for a total of 125 hours with several intermediate regrindings to improve the homogeneity of the samples. The sintering process was performed in the parallel processing system and consisted of the following heat treatment:

- heat to 935•C holding for 48 hours; then and
- cool to 500•Cand hold for 18 hours; then
- cool to 400•Cand hold for 10 hours; then
- cool to 300•Cand hold for 10 hours; then
- Cool to room temperature.

All sintering cooling processes were at the natural cooling rate of the oven. Resistance measurements were performed on the pellets and then shaped rods and bars were made from the pellets. Magnetization, both ZFC and FC data, were performed on the rods and resistivity measurements were performed on the bars.

MEASUREMENTS & RESULT ANALYSIS

A comparison of the dc magnetization, the ZFC and FC curves, is given in Figure 1. The Meissner fractions are tabulated in Table 1 along with the transition widths calculated (as described earlier) from the ac susceptibility (Figure 2), and resistivity (Figure 3) measurements. A characteristic comparison (10% Ca) for the Ca substitution series of all three measurements is given in Figure 4. The Ca samples also exhibit the small (< 3%) $BaCuo_2$ impurities that was seen in the Pr X- ray measurements.

Table 1: General Characteristics of Ca Substituted Y (20%Pr) BCO. A Number in Parenthesis Indicates That This Value is an Estimate

Ca Concentration	^{18}O Concentration	Meissner Fraction	Transition Width (K)		
(at.%)	(at.%)	@0.05Oe	Mag.	Susc.	Res.
0	(85)	35%	5.2	3.7	2.5
5	84	16%	2.0	1.4	1.3
10	78	12%	1.0	0.9	0.8
15	77	17%	1.9	1.5	1.0
20	76	21%	2.0	1.6	1.0
25	75	24%	1.7	1.4	1.3

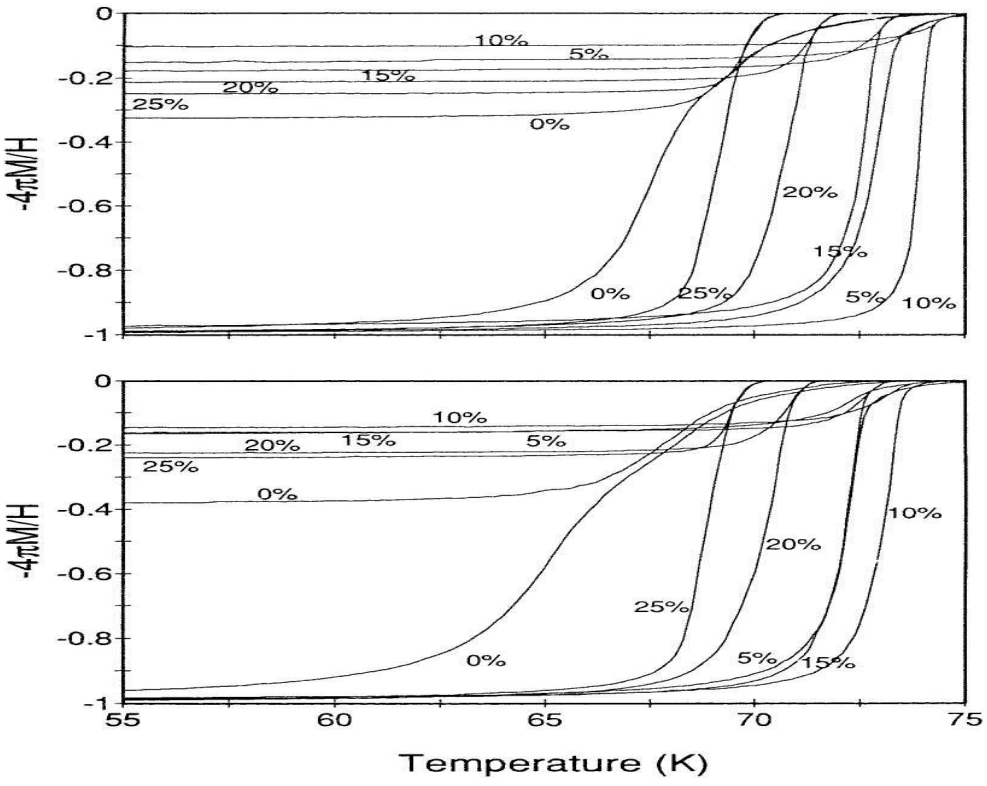

Figure 1: DC Magnetization vs. Temperature for Ca Substituted Y (20% Pr) BCO. Top ^{16}Odata; Bottom ^{18}O Data boththe ZFC and FC Data Are Shown Giving the Meissner Fraction

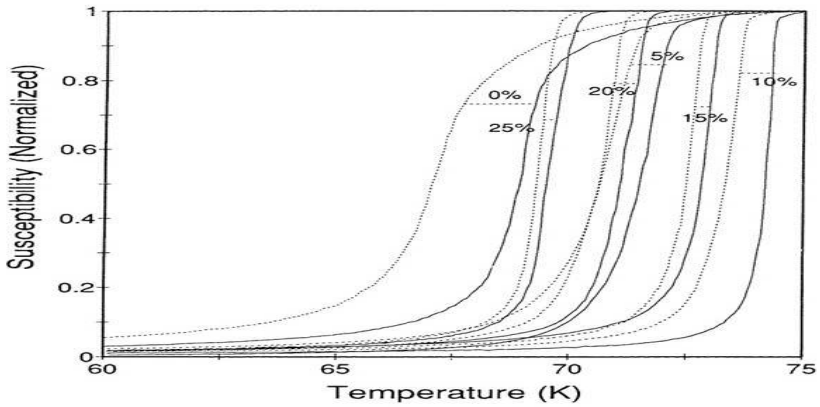

Figure 2: AC Susceptibility vs. Temperature for Ca Substituted Y (20% Pr) BCO. Solid Lines Represent The ^{16}O Isotope Data and the Dashed Lines Represent the^{18}o Isotope Data. A Dashed Lines Represent the ^{18}O Pairs is to Help Distinguish the Curves

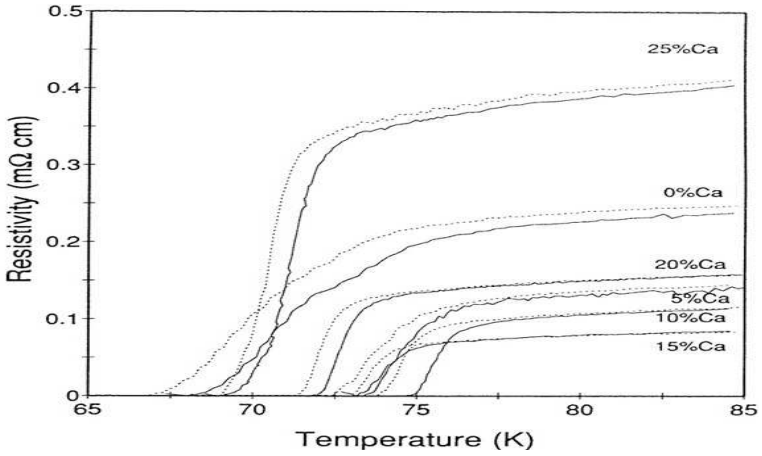

Figure 3: DC Resistivity vs. Temperature for Ca Substituted Y (20% Pr) BCO. The Solid Lines Represent the ^{16}O Isotope Data and the Dashed Lines Represent the ^{18}O Isotope Data

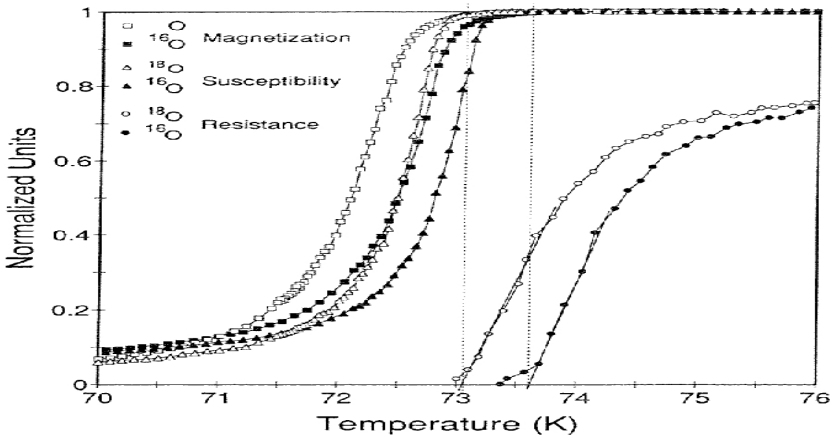

Figure 4: Comparison of Dc Magnetization, Ac Susceptibility and Resistance Measurements for $Y_{0.8}Pr_{0.2}Ba_2Cu_3O_{7-\Delta}$

The symbols represent the data. The solidfines provide continuity from point to point. The dotted lines are a visual aid forthe critical temperature of the resistive transition (obtained from the straight, line extrapolation). The resistive transition was normalized to R(85K).The obvious effect of the addition of Ca is an increase in the sharpness of the transition, and the elimination of the long tails at very small shielding. The width of the transition sharpness for all three

measurements with the additions of Ca and remains fairly constant with the increasing Ca concentration. As with the Pr, the resistance has the narrowest transition and the for dc magnetization has the widest. The isotopic shift measured is similar for both magnetic measurement but the resistivity measurement give a somewhat larger value for the higher concentration the size of the isotopic shift is affected also by the addition of Ca as it is reduced from that of the 20% Pr sample fabricated under the same conditions. The reductions is considerable since the change in Tc is quite small only about 7K throughout the entire substitution range The increase in Ca concentration in the sample has an effect on a number of properties of this system. First, Tcincrease then decrease with a maximum at approx. 10% Ca. Second, the Meissner fraction decrease then increase with a minimum at the 10% Ca. The normal state resistivity (at T=85K) is also parabolically dependent on the Ca concentration although its minimum is the slightly higher concentration. The parabolic effect in Tc (Figure 5) has been reported earlier The Tc data and the corresponding concentration of both the Pr, x, and the Pr:Ca, y, series has been fitted to the function proposed by Neumeier et al[59]. The dotted line in Figure 5 indicates this fit to equation [2.1], which gives$T_c(x,y)$ = 93.6K - (157K)(0.091-0.874x+y)^2-(93.3K)x [1]It should be noted that the last in equation [1], the linear pair-Breaking term, is an approximation to the more complicates digamma function which

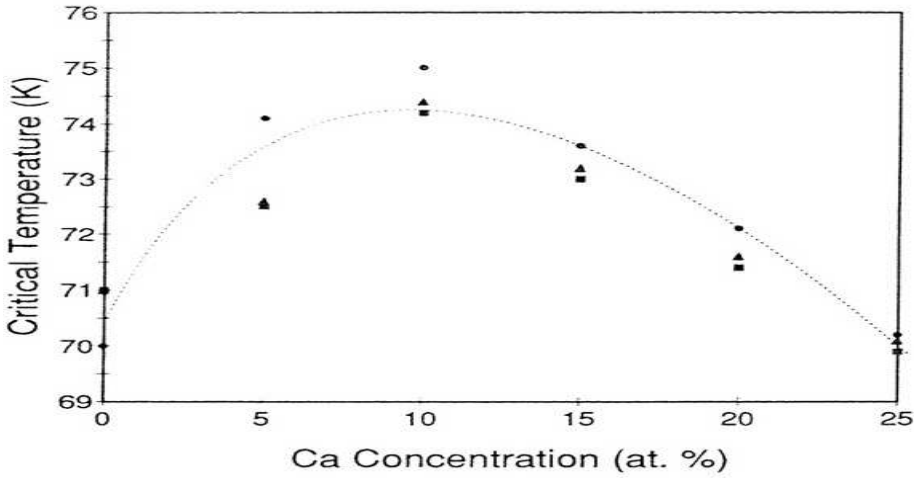

Figure 5: Critical Temperature vs. Ca Concentration.

The parabolic line is a fit of the data to the function[73] $T_c(x.y) = T_{co}$-A(γ-βx-y)^2-Bx,where x is the Pr concentration and y is the Ca concentration. We obtain T_{co} = 93.6K, a=157K, B= 93.3K, γ=0.091, and β=0.874. Both the Pr and the Pr:Ca data is used in fitting the function.should be used when dealing with the highest Pr concentrations because the pair-breaking here is no longer linear. The data, the Simplex fit to the data, and the computed isotope shift for the system of ($Y_{0.8}Pr_{0.2}Ba_2Cu_3O_{7-\delta}$), y = 0, 0.05, 0.1, 0.15,0.2,0.25, is given in Figures 6 to 10 respectively. The critical temperatures, the isotope shifts, and the computed values of the isotope coefficient for this system are given in Table 2. The relationship between α and ΔT_c as a function of T_c is given in Figure 10. Due to the smaller temperature range plotted here, the uncertainty in the temperature values appear large. In fact, they are actually smaller than that obtained in the Pr series. The value of T_c reported in the table is that which has been determined from the linear extrapolation technique as explained earlier. The isotopic shift, ΔT_c and the value of the isotope coefficient, α, change little over the short spread in critical temperature. The relationship between the critical temperature and α or ΔT_c is not clear. Considering the parabolic nature of the other properties of the system, a possible parabolic fit to α and ΔT_c, can be detected in the data. However, the large relative uncertainties also leave the possibility that of a linear relationship or perhaps even no change in ΔT_c at all. The empty symbols represent the ^{18}O data and the filled symbols represent the ^{16}O data the solid line represents the Simplex fit

to the data. The "+" symbols represent the shift, ΔTi, as a function of temperature, between the two fits to the data. The arrows on the magnetization and susceptibility graphs indicate the critical temperature obtained from a hear extrapolation of the bulk of the transition: 71.0k for both the magnetization and susceptibility. The upper limit for T_c is 76.0 K. The vertical dotted line (magnetic measurements) indicates the cutoff in the calculation of the mean of the isotope shift. The horizontal line indicates this mean, and its value is indicated above the line. The resistance measurements are normalized to R(85K). Not all temperature scales are equivalent.

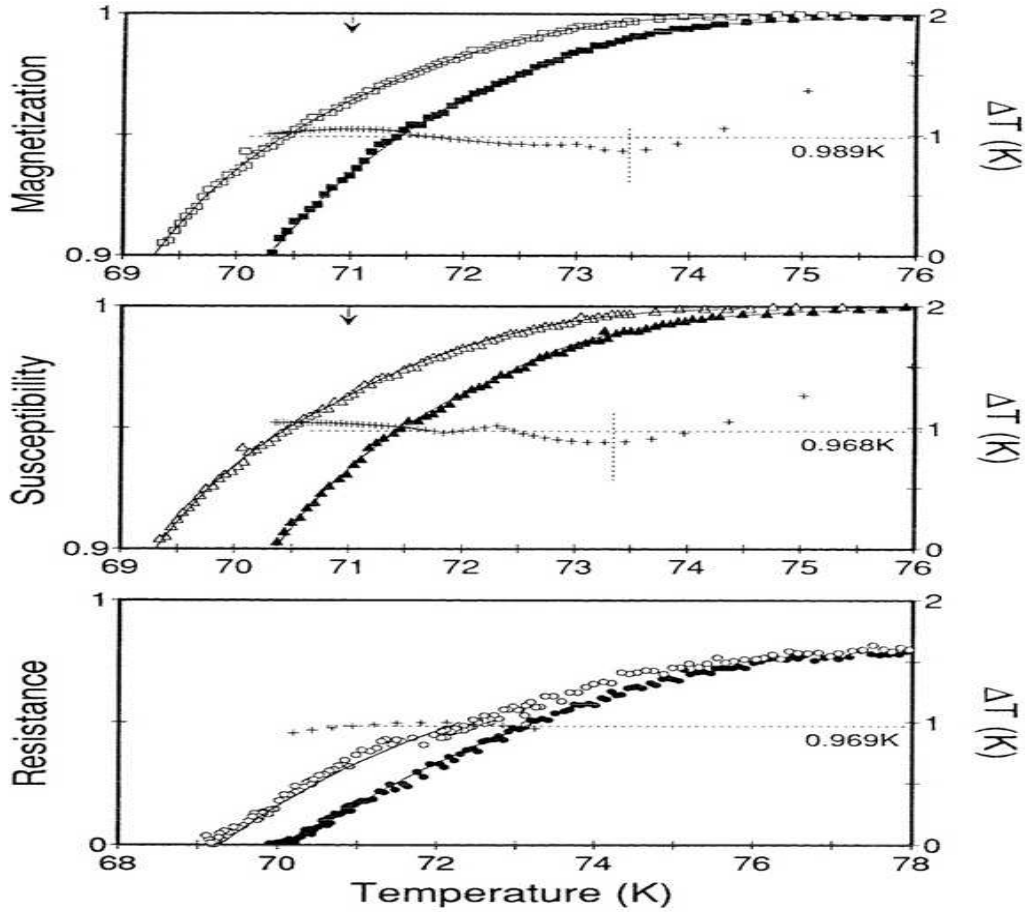

Figure 7: Oxygen Isotope Effect in $(Y_{0.8}Pr_{0.2}Ca_{0.05})Ba_2Cu_3O_{7-\Delta}$5% Ca, 20% Pr Substitution

The empty symbols represent the ^{18}O data and the filled symbols represent the^{16}Odata. The solid line represents the Simplex fit to the data. The "+" symbols represent the shift, ΔT_i, as a function of temperature, between the two fits to the data. The arrows on the magnetization and susceptibility graphs indicate the critical temperature obtained from a linear extrapolation of the bulk of the transition: 72.5K for the magnetization (upper limit = 74.8K) and 72.6K for the susceptibility (upper limit = 74.6K). No cutoff was imposed in the calculation of the mean of the isotope shift. The horizontal line indicates this mean, and its value is indicated above the line. The resistance measurements are normalized to R(85K). Not all temperature scales are equivalent.

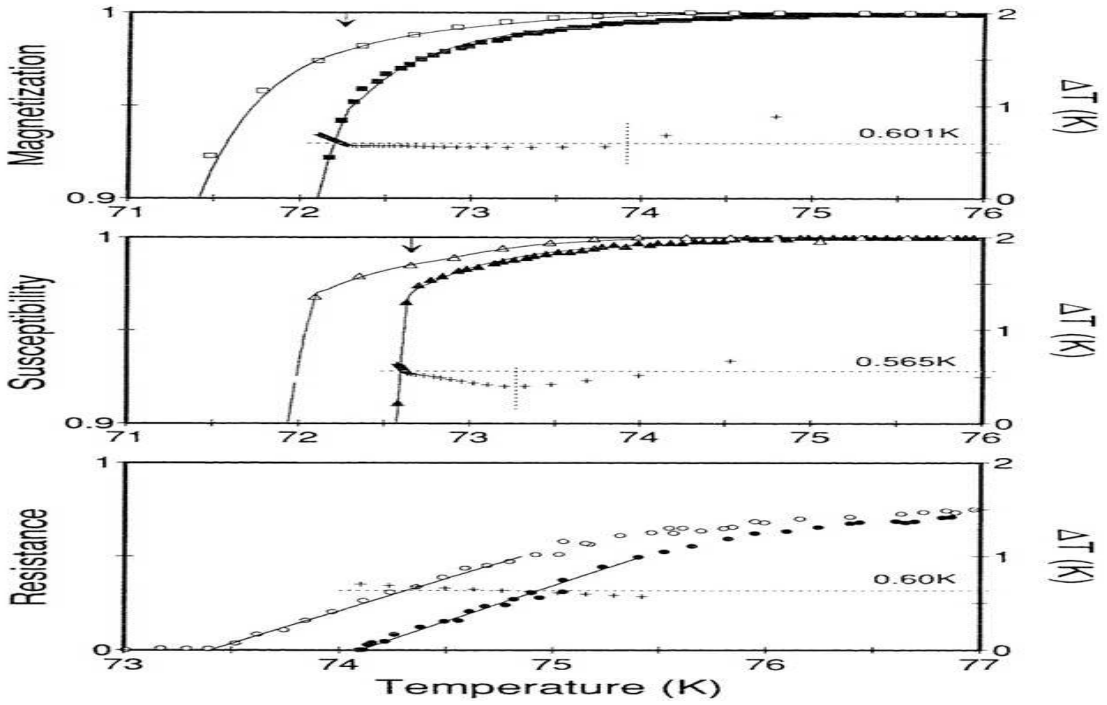

Figure 8: Oxygen Isotope Effect in ($Y_{0.8}Pr_{0.2}Ca_{0.1}$) $Ba_2Cu_3O_{7-\Delta}$), 10% Ca, 20% Pr Substitution

The empty symbols represent the ^{18}O data and the filled symbols represent the 16O data. The solid line represents the Simplex fit to the data. The "+" symbols represent the shift, ΔTi, as a function of temperature, between the two fits to the data. The arrows on the magnetization and susceptibility graphs indicate the critical temperature obtained from a linear extrapolation of the bulk of the transition: 74.2K for the magnetization (upper limit = 75.8K) and 74.4K for the susceptibility (upper limit = 75.7K). No cutoff was imposed in the calculation of the mean of the isotope shift. The horizontal line indicates this mean, and its value is indicated above the line. The resistance measurements are normalized to R(85K). Not all temperature scales are equivalent.

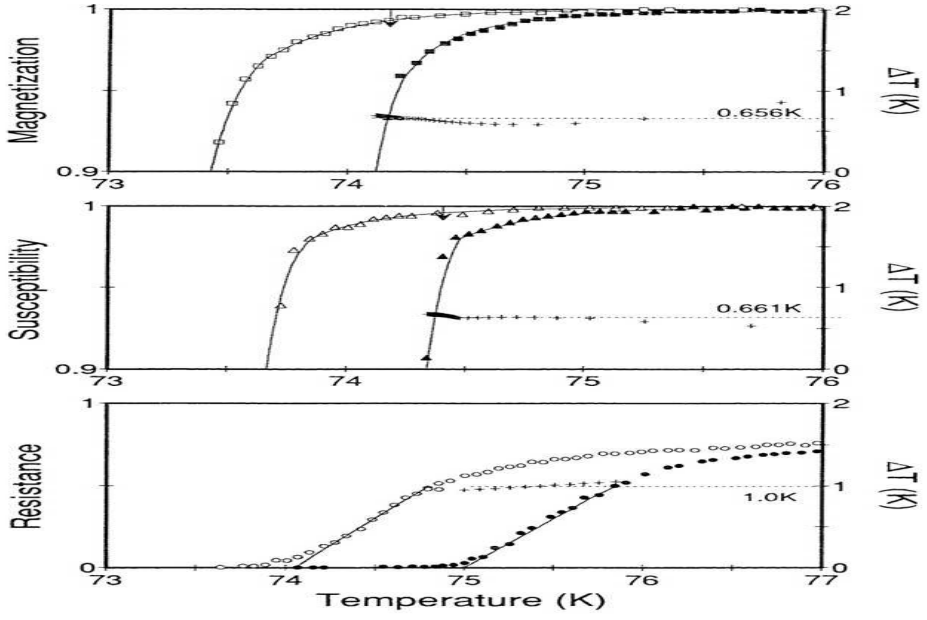

Figure 9: Oxygen Isotope Effect in ($Y_{0.65}Pr_{0.2}Ca_{0.15}$) $Ba_2Cu_3O_{7-\Delta}$), 15%Ca, 20% Pr Substitution

The empty symbols represent the ^{18}O data and the filled symbols represent the ^{16}O data. The solid line represents the Simplex fit to the data. The "+" symbols represent the shift, ΔTi as a function of temperature, between the two fits to the data. The arrows on the magnetization and susceptibility graphs indicate the critical temperature obtained from a linear extrapolation of the bulk of the transition: 73.OK for the magnetization (upper limit = 74.0K) and 73.2K for the susceptibility (upper limit = 73.8K). No cutoff was imposed in the calculation of the mean of the isotope shift. The horizontal line indicates this mean, and its value is indicated above the line. The resistance measurements are normalized to R(85K). Not all temperature scales are equivalent.

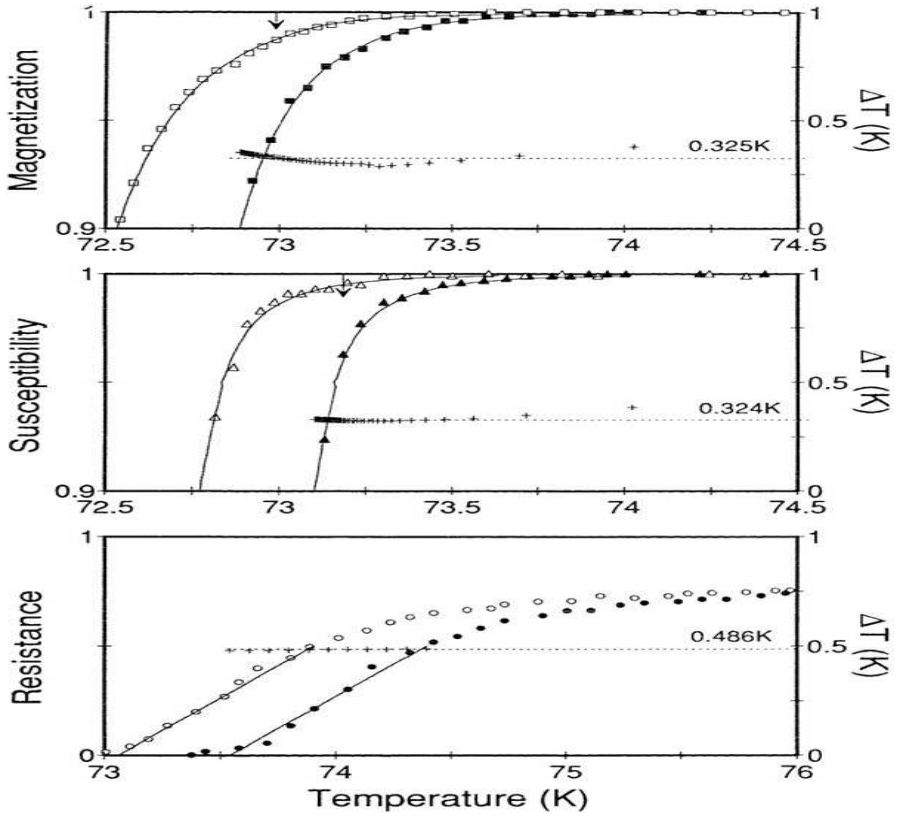

Figure 10: Oxygen Isotope Effect in ($Y_{0.6}Pr_{0.2}Ca_{0.2}Ba_2Cu_3O_{7-\Delta}$), 20% Ca, 20% Pr Substitution

The empty symbols represent the ^{18}O data and the filled symbols represent the ^{16}O data. The solid line represents the Simplex fit to the data. The "+" symbols represent the shift, ΔT_i, as a function of temperature, between the two fits to the data. The arrows on the magnetization and susceptibility graphs indicate the critical temperature obtained from a linear extrapolation of the bulk of the transition: 71.4K for the magnetization (upper limit = 72.54 and 71.6K for the susceptibility (upper limit = 72.6K). No cutoff was imposed in the calculation of the mean of the isotope shift. The horizontal line indicates this mean, and its value is indicated above the line. The resistance measurements are normalized to R(85K). Not all temperature scales are equivalent.

CONCLUSIONS

The fit of Tc to the concentrations of Pr, x, and Ca, y, to equation [2.1] fit very well. The maximum obtainable T_{co} (in the YBCO system) from our calculations is 93.6K, which appears to be a more reasonable value than Neumeier's 97K[59].We obtain an effective valence of +3.874 for the Pr, slightly smaller than Neumeier's +3.95,which is consistent with

the measurements that indicate a mixed valence between +3 and +4 With the optimum hole concentration, γ (= 0.091) being slightly smaller than Neumeier's (γ = 0.1), the predicted (relative) change in mobile hole concentration will also be very similar. In fact, only one parameter is significantly different from Neumeier's, and that is the coefficient A. Our value of A = 157K is 37% of Neumeier's A = 425K. The smaller value of A suggests that T_c would be more strongly affected by pair- breaking than by a change in hole concentration. The 20% Pr, 0% Ca sample made for comparison, is quite different from that of the Pr 20-50% system. Its T_c is about 5K lower than before, the magnetic transition width is much broader (-1.4X) and its isotope shift is also larger (-1.5X). Clearly the preparation technique greatly affects the characteristics and hence the quality of the samples. As the highest quality materials, single crystals, tend to have very sharp transitions, and the polycrystalline materials have broader transitions, the relationship between the isotope shift and the width of the transition was investigated for both the Pr and Pr:Ca substituted materials. A linear correlation between the ΔT_c and the transition width is found. The intercept for a zero-width transition remains finite, in the range of 0.3K to 0.4K. This result poses the question of whether the isotope shift should actually be taken as a constant for this system.

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