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THE OXYGEN ISOTOPE EFFECT IN Pr, Ca, AND Zn SUBSTITUTED YBa₂Cu₃O_{7-δ} AND EuBa₂Cu₃O_{7-δ}.J.P. FRANCK^a, S. GYGAX^b, G. SOERENSEN^b, E. ALTSHULER^b, A. HNATTW^a, J. JUNG^a, M.A.-K. MOHAMED^a, M.K. YU^a, G.I. SPROULE^c, J. CHRZANOWSKI^b AND J.C. IRWIN^b.^a Department of Physics, University of Alberta, Edmonton, Alberta, Canada, T6G 2J1^b Department of Physics, Simon Fraser University, Burnaby, B.C., Canada V5A 1S6^c Institute for Microstructural Sciences, National Research Council, Ottawa, Canada K1A 0R9

The oxygen isotope effect α was studied for Pr and Ca substituted YBCO, and for Zn substituted EuBCO and YBCO. α increases to values near 0.5 for the highest Pr concentrations, and lowest transition temperatures. In the Zn substituted system α remains small to the lowest transition temperatures. It is shown that the isotope exponent is a function of mobile hole concentration, it decreases with increasing hole concentration and remains low for over-doping.

We are reporting determinations of the oxygen isotope effect in Pr, Ca and Zn substituted YBCO. We prepared the series $(Y_{1-x}Pr_x)Ba_2Cu_3O_{6.92}$ for $x = 0.2, 0.3, 0.4,$ and $0.5,$ and the series $(Y_{1-x-y}Pr_xCa_y)Ba_2Cu_3O_{6.92}$ for $x = 0.2,$ and $y = 0.05$ to $0.30,$ and for $y = 0.05$ and $x = 0.2, 0.3$ and $0.4.$ The samples were prepared from the high purity oxides; calcining for extended periods between 125 and 620 hours was used with frequent regrinding. Three identical pellets of calcined material were parallel-processed in flowing, and purified $^{16}O_2$ or $^{18}O_2.$ This method has been described previously¹. The sintering conditions were designed to ensure full oxygenization, we determined the total oxygen content as 6.92 ± 0.03 by iodometric titration. The ^{18}O concentration was determined by SIMS and found for different samples between 75 and 94.3%. Raman scattering on several samples agreed with this. We also prepared samples of $EuBa_2(Cu_{1-x}Zn_x)_3O_{7-δ}$ for $x = 0.015, 0.025,$ and $0.035,$ and one sample of $YBa(Cu_{0.94}Zn_{0.06})_3O_{7-δ}.$

The isotopic shift was observed resistively and magnetically by low field dc magnetization and ac susceptibility. We find in general good agreement between both methods, but sometimes the magnetically determined shifts are larger than the resistive ones. In Fig. 1 we show the oxygen isotope exponent $\alpha = -d \ln T_c / d \ln m,$ normalized to 100% ^{18}O concentration, as a function of transition temperature. For the system $(Y_{1-x}Pr_x)Ba_2Cu_3O_{6.92}$ α increases to values near 0.5 at the

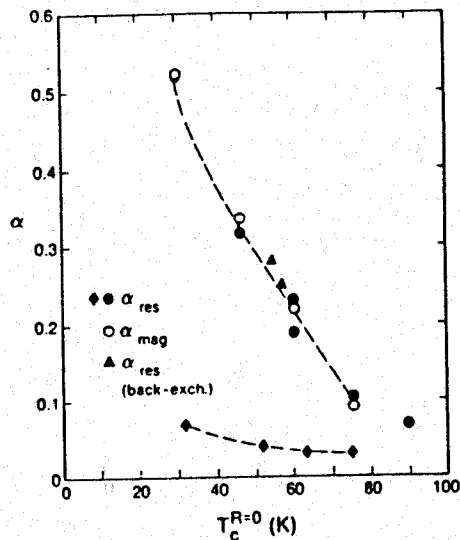


FIGURE 1

The normalized oxygen isotope exponent α (100% ^{18}O) as a function of transition temperature $T_c.$ Round symbols refer to $(Y_{1-x}Pr_x)Ba_2Cu_3O_{6.92}, x = 0.2, 0.3, 0.4$ and $0.5.$ Diamonds refer to $EuBa_2(Cu_{1-x}Zn_x)_3O_{7-δ}, x = 0.015, 0.025, 0.035,$ and to $YBa_2(Cu_{0.94}Zn_{0.06})_3O_{7-δ}$ (transition temperature 32 K). Open symbols: magnetic determinations, closed symbols: resistive determinations.

highest Pr concentration². Ca substitution at constant Pr content sharpens the transition, T_c goes through a shallow maximum and α decreases somewhat. This is shown in Fig. 2. For the Zn substituted systems, the isotope

exponent remains small, and only shows a slight increase to $\alpha = 0.07$ for $T_c = 32$ K. It is clear from Fig. 1 that the transition temperature does not determine the size of the isotope exponent.

In the Pr and Ca substituted series of YBCO it is believed that the transition temperature is influenced by two effects³: Magnetic pair-breaking due to Pr, and changes in mobile hole concentration; Pr reduces the mobile hole concentration with an effective valence near 4 and Ca increases it^{3,4}. We determined the mobile hole concentration by using the fitting procedures of Neumeier et al³, the fits to the transition temperature in Fig. 2 are obtained in this way. The relative mobile hole concentrations Δp obtained in this way allowed us to plot both T_c and α as function of the mobile hole concentration per CuO sheet, Fig. 2. The isotope exponent as function of mobile hole concentration Δp fits a single curve, it increases for low Δp , and decreases for larger values, and for over-doping ($\Delta p > 0$).

The substitution of Zn also leads to a rapid decrease in transition temperature. Various explanations have been advanced: Magnetic pair breaking⁵ or increased hole concentration beyond the optimal one⁶. Hall effect measurements definitely show an increase in mobile hole concentration⁶ and we have used the analysis of ref. (6) to estimate this concentration for our Zn substituted samples. We see from Fig. 2 that the low isotope exponent in this system fits the general relation between α and Δp in spite of the drastically different behaviour of T_c . It appears therefore that the mobile hole concentration per CuO plane is the dominating parameter for the size of the isotope exponent.

A number of theoretical investigations have predicted a low isotope effect near the maximum T_c in a series, based on van Hove singularities in the carrier density of states^{7,8,9} near the Fermi surface. Our data are in general agreement with this.

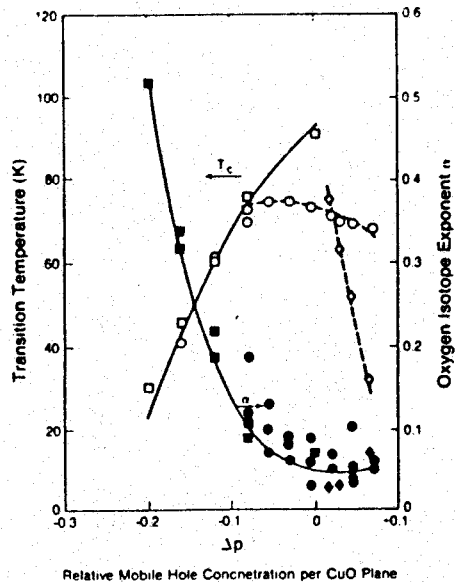


FIGURE 2
The transition temperature T_c (open symbols) and normalized oxygen isotope exponent α (filled symbols) as a function of relative mobile hole concentration per CuO plane. Squares refer to the system $(Y_{1-x}Pr_x)Ba_2Cu_3O_{6.92}$, circles to $(Y_{1-x-y}Pr_xCa_y)Ba_2Cu_3O_{6.92}$, and diamonds to $EuBa_2(Cu_{1-x}Zn_x)_3O_{7.8}$ and $YBa_2(Cu_{1-x}Zn_x)_3O_{7.8}$. The fit to the isotope exponent α is only a guide to the eye.

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