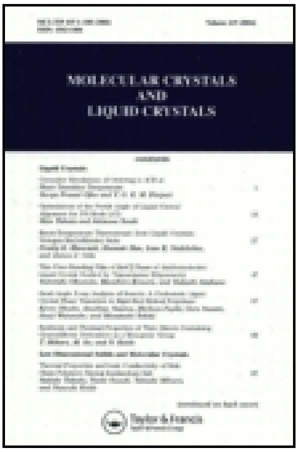

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An Effect of Substitution on the Superconductive Property in Perovskite-Like Oxides

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AN EFFECT OF SUBSTITUTION ON THE SUPERCONDUCTIVE PROPERTY IN PEROVSKITE-LIKE OXIDES

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Abstract The critical temperature, Tc, of $YBa_2Cu_3O_{7-\delta}$ was lowered by the partial substitution of Nb, Mo, Al, Zr, Bi, Sn, Ti and In for Cu. With a decrease in Tc, lattice parameter a increased, but lattice parameter b was almost constant. The superconductive property of $Bi_2Sr_2CaCu_2O_{9-\delta}$ samples was significantly affected by the preparation condition and by cation substitution. With an increase in Gd substitution for Ca, the oxygen content increased, which also accompanied a decrease in Tc. The lattice parameter c decreased with an increase in the oxygen content. In various $Bi_2Sr_2CaCu_2O_{9-\delta}$-based samples, the critical temperature tended to increase with an increase in lattice parameter c.

INTRODUCTION

The superconducting properties of oxides with perovskite-like crystal structures are significantly affected by the nonstoichiometry of oxygen in the solid or by substitution of metal cations. Substitution of several rare earth metals for yttrium do not affect critical temperature, Tc, of $YBa_2Cu_3O_{7-\delta}$ the superconductive properties.[1] But substitution of Sr or La for Ba reduces Tc.[2,3] The critical temperature was in many cases lowered with substitution for Cu.[4-7] Xiao et al.[8] have investigated the systematic change in Tc with substitution of 10 atomic % of Cu for 3d metals from Ti to Zn.

The substitution effect in Bi-Sr-Ca-Cu-O system is more complicated, but Pb-substitution for Bi is very effective in producing the high Tc phase.[9] The dope of rare earth metals at Ca site generally gives rise to low Tc value.[10-12] In the present study, we investigated the effect of metal substitution on superconducting properties of $YBa_2Cu_3O_{7-\delta}$ and $Bi_2Sr_2CaCu_2O_{9-\delta}$. The relation among Tc, lattice constants, and oxygen nonstoichiometry has been discussed.

Experimental

The samples were prepared by the solid state reaction of component oxides. The oxide or carbonate powders were ball-milled overnight and heated at 800-900 °C in air. The pulverized samples were pressed into pellets (10 mm x 2 mm) and finally sintered for 5 h at 950 °C for $YBa_2Cu_3O_{7-\delta}$ or 800 °C for $Bi_2Sr_2CaCu_2O_{9-\delta}$ systems.

The resistivity and inductance measurements were carried out in a temperature range of 10 K to room temperature. The resistivity was measured by dc-4 probe method. Meissner effect was roughly estimated from the inductance change of a coil with temperature, in which a sample capsule is placed. The content of oxygen and the average valence of Cu ion in the samples were estimated by iodometry. The valence of Bi was assumed as tervalent.

RESULTS AND DISCUSSIONS

Effect of substitution for Cu in $YBa_2Cu_3O_{7-\delta}$ system

Temperature dependence of resistivity and magnetic susceptibility of $YBa_2Cu_3O_{7-\delta}$ are shown in Figure 1. The resistivity steeply decreased at 97 K and reached to zero at 94 K. The magnetic susceptibility also decreased at 94 K due to the Meissner effect. Several metals (Nb, Mo, Al, Sb, Zr, Bi, Sn, Ti, and In were substituted for 1 atomic% of Cu in $YBa_2Cu_3O_{7-\delta}$. The lattice parameters of the substituted sample, $YBa_2(Cu_{0.99}A_{0.01})_3O_{7-\delta}$ are plotted as a function of Tc (Figure 2). The relation between Tc and lattice constant c was unclear, but lattice parameter b

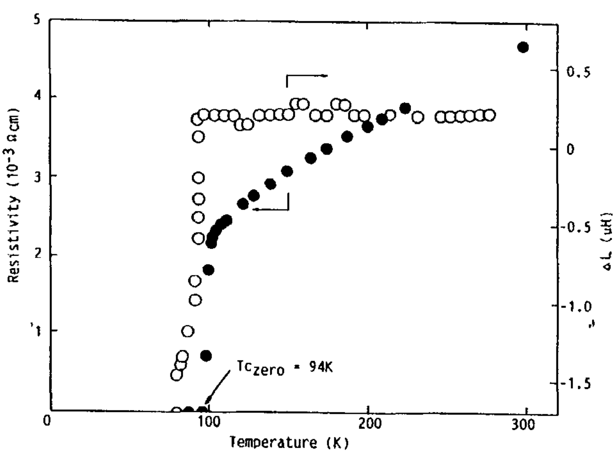

FIGURE 1 Temperature dependence of resistivity and magnetic susceptibility for $YBa_2Cu_3O_{7-x}$.
$L = L_{sample} - L_{blank}$

was unchanged among the samples. It is noted that lattice parameter a increased with decreasing Tc. The lattice parameters a and b, corresponding to the Cu-O-Cu bond distance, are expected to increase with a removal of oxide ion from the lattice due to electrostatic repulsion between neighboring Cu ions. Thus the relation between lattice parameter a and Tc appears to be related with oxygen deficiency along the a direction.

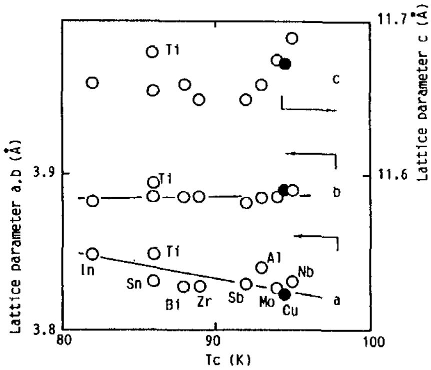

FIGURE 2 Relationship between Tc and lattice parameter of $YBa_2(Cu_{0.99}A_{0.01})_3O_{7-x}$ system.
(A ; Nb,Mo,Al,Sb,Zr,Bi,Sn,Ti,In)

<u>Effect of Gd-substitution for Ca in $Bi_2Sr_2CaCu_2O_{9-\delta}$</u>

The Bi-Sr-Ca-Cu-O system consists of two superconducting phases, which are the low Tc phase (Tc = 80 K) and the high Tc phase (Tc = 110 K). Preparation of the sample with the single low Tc phase is much easier than that with the single high Tc phase. A variety of metals were substituted in this system and the effect on superconducting properties was investigated. Figure 3 shows the effect of Gd-substitution for Ca on temperature dependence of resistivity. The critical temperature Tc was lowered with increasing Gd content. The relation between oxygen content and lattice parameters is shown in Figure 4. The oxygen content increased with increasing Gd-content, accompanying the reduction of Tc. Lattice parameter a remained constant, whereas lattice parameter c decreased with an increase in oxygen content.

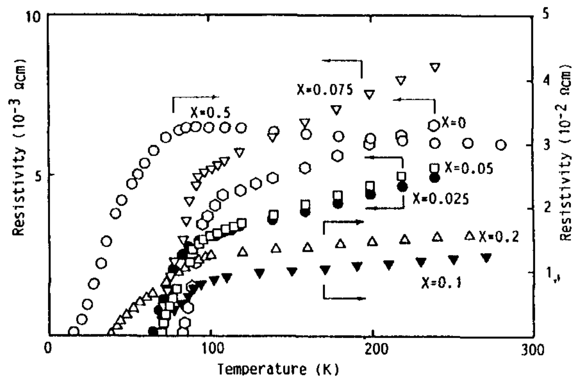

FIGURE 3 Temperature dependence of resistivities for $Bi_2Sr_2Ca_{1-x}Gd_xCu_2O_{9-\delta}$ system.

Thus the oxygen occupancy is preferencially changed along the c direction relative to a direction. The superconductive properties of this system are strongly affected by oxygen nonstoichometry.

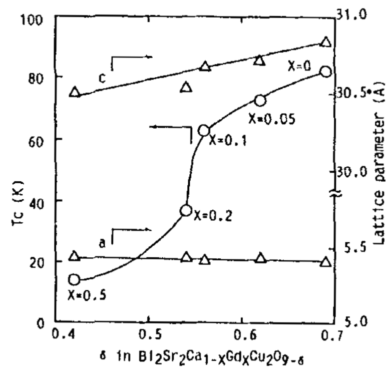

FIGURE 4 Relation of Tc and lattice parameter with value in $Bi_2Sr_2Ca_{1-x}Gd_xCu_2O_{9-\delta}$ system.

__The relation between Tc and the lattice constant in cation substituted Bi-Sr-Ca-Cu-O system__

The effects of preparation condition of $Bi_2Sr_2CaCu_2O_{9-\delta}$ and of various substituents are investigated in this section. Figure 5 summarizes the relation between lattice parameters and Tc in cation substituted or un-substituted $BiSrCaCuO_3$. The lattice constant a was unchanged

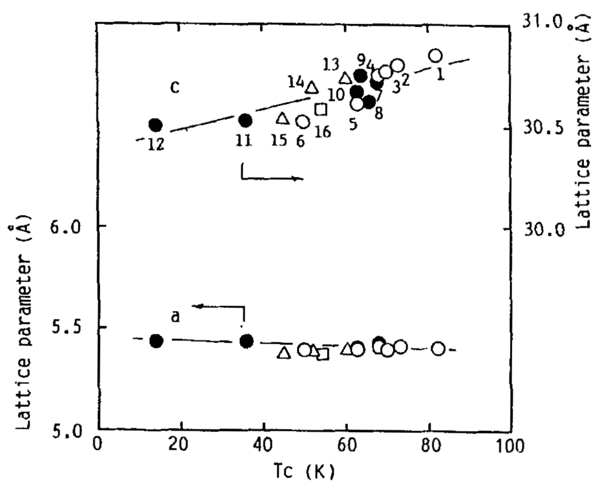

FIGURE 5 Relation of lattice parameter with Tc in $Bi_2Sr_2CaCu_2O_{9-\delta}$ system.

1; $Bi_2Sr_2CaCu_2O_{9-\delta}$ (air liq.N_2 quench) 2; $Bi_2Sr_2CaCu_2O_{9-\delta}$ (N_2)
3; $Bi_2Sr_2CaCu_2O_{9-\delta}$ (air 20°C quench) 4; $Bi_2Sr_2CaCu_2O_{9-\delta}$ (N_2 20°C quench)
5; $Bi_2Sr_2CaCu_2O_{9-\delta}$ (air) 6; $Bi_2Sr_2CaCu_2O_{9-\delta}$ (O_2)
7; $Bi_2Sr_2Ca_{0.95}Gd_{0.05}Cu_2O_{9-\delta}$ 8; $Bi_2Sr_2Ca_{0.925}Gd_{0.075}Cu_2O_{9-\delta}$
9; $Bi_2Sr_2Ca_{0.075}Gd_{0.025}Cu_2O_{9-\delta}$ 10; $Bi_2Sr_2Ca_{0.9}Gd_{0.1}Cu_2O_{9-\delta}$
11; $Bi_2Sr_2Ca_{0.8}Gd_{0.2}Cu_2O_{9-\delta}$ 12; $Bi_2Sr_2Ca_{0.5}Gd_{0.5}Cu_2O_{9-\delta}$
13; $Bi_2Sr_{1.8}Pb_{0.2}CaCu_2O_{9-\delta}$ 14; $Bi_2Sr_{1.9}Pb_{0.1}CaCu_2O_{9-\delta}$
15; $Bi_2Sr_{1.6}Pb_{0.4}CaCu_2O_{9-\delta}$ 16; $Bi_{1.9}Sb_{0.1}Sr_2CaCu_2O_{9-\delta}$

irrespective of Tc. It is noted that the lattice constant c decreased with decreasing Tc. The large c value corresponds to small interaction between the neighboring superconductive a-b planes.

CONCLUSION

The effect of cation substitution in high Tc superconductor is summerized as follows.

1) Lattice parameter b was unchanged with cation substitution for Cu in $Ba_2Cu_3O_{7-\delta}$, whereas the lattice constant a increased with a decrease in Tc.

2) In the series cation substituted $Bi_2Sr_2CaCu_2O_{9-\delta}$ samples, the decrease in Tc accompanies the decrease in lattice parameter c, but lattice parameter a remains constant. The superconductive properties are strongly influenced by oxygen nonstoichiometry.

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