



Mechanism of a strong rise of T_c due to the calcium doping in $Y_{1-x}Ca_xBa_2Cu_{2.8}Zn_{0.2}O_y$

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ABSTRACT

The normal-state transport properties and the critical temperature, T_c , for three series of $Y_{1-x}Ca_xBa_2Cu_{2.8}Zn_{0.2}O_y$ ($x = 0-0.2$) samples with different oxygen content have been investigated and discussed in comparison with results for other calcium-doped $YBa_2Cu_3O_y$. We have observed the calcium doping to restore the superconductivity in the $YBa_2Cu_3O_y$ system even in case of the preliminary T_c suppression by the direct destruction of the CuO_2 planes due to a preliminary doping by zinc. The T_c value rises strongly with increasing calcium content and this rise becomes faster for the oxygen-reduced series. Based on a narrow-band model, the parameters of the band spectrum structure and the charge-carrier system in the normal state were determined from the analysis of the thermopower temperature dependences in order to reveal the mechanism of the T_c rise under the Ca influence. It is shown that the T_c increase in $Y_{1-x}Ca_xBa_2Cu_{2.8}Zn_{0.2}O_y$ is related to the direct Ca ions effect. The mechanism of the calcium influence on the T_c value is discussed with respect to the Ca-induced conduction band modification. The calcium doping in $Y_{1-x}Ca_xBa_2Cu_{2.8}Zn_{0.2}O_y$ is shown to give a noticeable contribution to the density-of-states function leading to a strong rise in its value at the Fermi level that results in a restoration of the superconducting properties.

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1. Introduction

The effect of the calcium doping in the $YBa_2Cu_3O_y$ system including the case of double substitutions for samples with other dopants in different lattice positions attracts the great attention [1–4]. The specific effect of calcium doping is the ability of this impurity to restore the superconductivity suppressed by some other factors [4–8]. For example, additional Ca doping in $Y_{1-x}Ca_xBa_{2-z}La_zCu_3O_y$ and $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ with fixed La or Co content results in T_c increase or even the superconductivity restoration [4,5,7,8]. This effect is related to the fact that all the properties of the above systems were preliminary changed by the first dopant with a valence higher than the substituted cation one [5–8]. Such non-isovalent impurities lead to increasing oxygen content over its stoichiometric value and, as a consequence, to a disordering in the oxygen subsystem. On the other hand, it is well known that calcium in yttrium positions plays the role of an acceptor-like impurity (see, for example, Ref. [8] and references therein). Due to its lower valence relative to the valence of substituted yttrium, calcium doping leads to an increasing oxygen deficit [7–12], thus compensating the influence of the first impurity on the charge

balance in the lattice. For this reason, the oxygen content in the $Y_{1-x}Ca_xBa_{2-z}La_zCu_3O_y$ and $Y_{1-x}Ca_xBa_2Cu_{3-z}Co_zO_y$ systems with fixed z goes down with x towards its stoichiometric value $y \approx 7$ that induces the oxygen subsystem ordering and, as a result, an increase in the T_c value [7,8].

However, this effect of the oxygen subsystem ordering cannot be responsible for the recovery of superconducting properties observed in oxygen deficit $Y_{1-x}Ca_xBa_2Cu_3O_y$ samples [7,8,12,13]. In this case, according to our conclusions [7,8,12], increasing T_c is rather caused by the Fermi level, E_F , movement through the conduction band. In the oxygen-rich samples E_F shifts with x away from the maximum of the density-of-states, DOS, function that leads to a drop in T_c , while in the oxygen-reduced ones it shifts towards this maximum that results in a rise of T_c .

Thus, calcium restores the superconducting properties in $YBa_2Cu_3O_y$ with oxygen deficit, as well as in pre-doped $YBa_2Cu_3O_y$ with over stoichiometric oxygen content induced by non-isovalent impurities in different positions. Physical reasons for these similar effects are likely to be different and call for further studies. The investigation of the calcium effect in $YBa_2Cu_3O_y$ where the superconductivity was preliminarily reduced by other factors can help in elucidating the mechanism of an unusual Ca influence on the superconducting properties. In our opinion, the most interesting impurity for such a study is zinc in cooper positions. The oxygen deficit and $Co \rightarrow Cu$ or $La \rightarrow Ba$ substitutions affect the

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superconductivity indirectly not disturbing the CuO_2 planes. On the contrary, zinc immediately substitutes for the in-plane copper strongly suppressing T_c [14–17] due to an influence on the pairs formation in the CuO_2 planes responsible for the superconductivity. In addition, unlike $\text{Co} \rightarrow \text{Cu}$ and $\text{La} \rightarrow \text{Ba}$ substitutions zinc doping does not affect the oxygen content [14,15]. For the above reasons, the study of the properties of $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{2.8}\text{Zn}_{0.2}\text{O}_y$ samples could allow one to reveal the direct effect of the calcium doping and thus to understand the mechanism of this impurity influence on the superconductivity in $\text{YBa}_2\text{Cu}_3\text{O}_y$.

2. Samples and experimental details

The ceramic samples of $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{2.8}\text{Zn}_{0.2}\text{O}_y$ ($x = 0.0\text{--}0.2$) were prepared by the standard solid-state processing technique from the high-purity oxides and carbonates mixed in the required proportions. The mixtures were heated, reground, pressed into pellets and then sintered three times at $T = 900, 920$ and 930°C for the first, second, and third synthesis, correspondingly, with intermediate regrinding. Finally, the samples were annealed in flowing oxygen at $T = 450^\circ\text{C}$ for 2 h and slowly furnace cooled down to room temperature. By such a way, a series of as-prepared samples with a high oxygen content was prepared. Then the samples were additionally annealed in vacuum at $T = 460$ and 470°C for 2 h in order to create two additional series with an oxygen deficit. The oxygen content in Ca-free samples of those three series determined by the iodometric titration technique was equal to 6.88, 6.74, 6.69, correspondingly. X-ray-diffraction analysis (Siemens D-500 diffractometer, Cu K_α radiation) has shown that all the samples had a typical Y-123-like structure belonging to the orthorhombic crystal symmetry. Samples were almost of single phase with a small amount of foreign phases (not exceeding 1–2% according to the reflection intensity). The lattice parameters changed insignificantly with x in ranges of $a = 3.825\text{--}3.84 \text{ \AA}$, $b = 3.88\text{--}3.89 \text{ \AA}$, $c = 11.65\text{--}11.68 \text{ \AA}$.

The resistivity and thermopower were measured in the temperature range of $T = T_c \pm 300 \text{ K}$ for all the samples. The resistivity measurements were performed by the standard four-probe low-frequency ac (20 Hz) method. The thermopower was measured relative to copper electrodes and then calculated by correcting for the thermopower of copper. The temperature difference between the two ends of the sample was kept around 2 K throughout the measuring procedure.

3. Experimental results

The temperature dependences of the resistivity, ρ , demonstrate the usual for the $\text{YBa}_2\text{Cu}_3\text{O}_y$ system behavior. For all the as-prepared samples they are linear in the temperature range of $T = T_c - 300 \text{ K}$. In the oxygen-reduced series all the $\rho(T)$ curves demonstrate a semiconductor-like behavior. The variation of T_c (defined as the midpoint of the resistive superconducting transition) with calcium content is shown in Fig. 1. Here and below solid lines approximating the obtained results by eye are shown for indicating tendencies in variation of the corresponding parameter with increasing calcium content for each sample series. It is clearly seen, that for all three $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{2.8}\text{Zn}_{0.2}\text{O}_y$ series the T_c value rises almost linearly as calcium content increases. Vacuum annealing leads to a decrease in T_c for oxygen-reduced series, as a whole. Nevertheless, both these series demonstrate a more pronounced restoration of the superconductivity with increasing x than the as-prepared one. Thus, the T_c value in the studied compound rises with increasing calcium content, which is similar to the case of $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_{2-z}\text{La}_z\text{Cu}_3\text{O}_y$ and $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{3-z}\text{Co}_z\text{O}_y$ with fixed z . However, the physical reason for this phenomenon seems to be rather different since doping with Zn, in contrast to Co and La, does

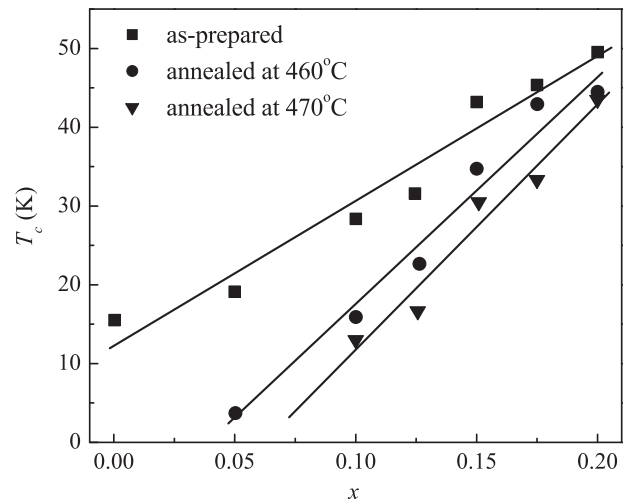


Fig. 1. Critical temperature vs. calcium content in three $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{2.8}\text{Zn}_{0.2}\text{O}_y$ series.

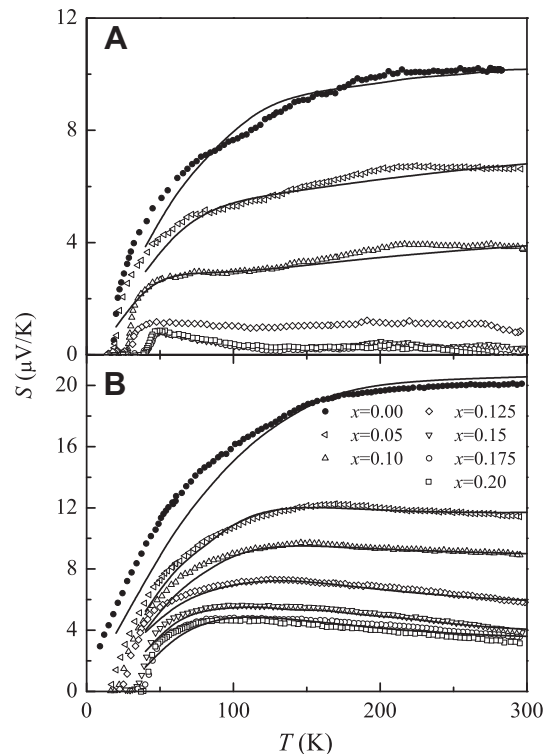


Fig. 2. Thermopower temperature dependences for $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{0.8}\text{Zn}_{0.2}\text{O}_y$. A – starting samples, B – samples annealed at $T = 460^\circ\text{C}$. Symbols are the experimental data, solid lines represent the results of calculations within the narrow-band model.

not lead to increasing y over its stoichiometric value. Thus, increasing x cannot lead to ordering of the oxygen subsystem.

Fig. 2 shows the thermopower temperature dependences for the as-prepared and one of the oxygen-reduced (annealed at 460°C) $\text{Y}_{1-x}\text{Ca}_x\text{Ba}_2\text{Cu}_{2.8}\text{Zn}_{0.2}\text{O}_y$ series. $S(T)$ curves for the series annealed at 470°C are analogous to the last case. Fig. 3 presents the variation of the thermopower value at $T = 300 \text{ K}$, S_{300} , for all the three series studied. One can see that increasing oxygen deficit induced by samples annealing leads to a rise of S_{300} for a sample series as a whole. The S_{300} value decreases with doping level for all the series within the range of $x = 0\text{--}0.15$ and becomes practically

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