Possible Inhomogeneity of Superconductivity in (Y,Ca)Ba$_2$Cu$_3$O$_{7-\delta}$ Probed by Magnetic Susceptibility and Specific Heat

S. Chamura, T. Adachi, Y. Tanabe and Y. Koike

Department of Applied Physics, Tohoku University, 6-6-05 Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan
E-mail: koike@teion.apph.tohoku.ac.jp (Y. Koike)

Abstract. In order to investigate the possible inhomogeneity of superconductivity from measurements reflecting bulk properties of a sample, we have measured the magnetic susceptibility, $\chi$, and specific heat of slightly overdoped and underdoped $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ polycrystals. The absolute value of $\chi$ at 2 K, $|\chi_2K|$, on field cooling reflecting the superconducting (SC) volume fraction has been found to decrease monotonically with decreasing hole-concentration. From the specific heat measurements, on the other hand, it has been found that the so-called Sommerfeld constant is not zero but has a finite value even in the SC state. These results are quite similar to those observed in La$_2$-$x$Sr$_x$CuO$_4$ and suggest that a phase separation into SC and normal-state regions takes place in $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ as well. It appears that the phase separation is a generic feature in high-$T_c$ cuprates.

1. Introduction

The microscopic electronic inhomogeneity in high-$T_c$ cuprates suggested from scanning-tunneling-microscopy experiments has attracted much attention in recent years [1]. In the overdoped regime of high-$T_c$ cuprates, experimental results using probes reflecting bulk properties of a sample, such as specific heat [2,3], $\mu$SR [4,5], NMR [6], magnetic susceptibility, $\chi$ [7,8], have supported this inhomogeneous picture. In the underdoped regime of high-$T_c$ cuprates, on the other hand, specific heat measurements have revealed that the so-called residual Sommerfeld constant in the ground state, $\gamma_0$, is not zero in superconducting (SC) samples of La$_{2-x}$Sr$_x$CuO$_4$ (LSCO) [9,10], suggesting the presence of normal-state regions in SC samples and therefore supporting the inhomogeneous picture.

Formerly, we measured both $\chi$ on field cooling and the specific heat in the underdoped and optimally doped LSCO in order to investigate the possible inhomogeneity of superconductivity [11,12]. It has been found that both the Meissner volume fraction regarded as corresponding to the SC volume fraction in a sample and the $\gamma_0$ value exhibit significant $x$-dependences suggesting the occurrence of a phase separation into SC and normal-state regions in the underdoped regime, though the existence of the so-called stripe correlations of spins and holes [13] might affect the results.

In this paper, we have investigated the possible phase separation into SC and normal-state regions in $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$, in which effects of the stripe correlations are considered to be weak, by means of both $\chi$ and specific heat measurements.
2. Experimental
Polycrystalline samples of $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ were prepared by the solid-state reaction method [14]. The hole concentration, $p$, of a sample was changed by annealing it in air and was estimated using an empirical formula relating to the value of the thermoelectric power at 290 K and the $p$ value [15]. The $\chi$ measurements were performed down to 2 K using a SQUID magnetometer. The specific heat was measured by the thermal relaxation method down to 2 K using a commercial apparatus (Quantum Design, PPMS).

3. Results and discussion
Figure 1 shows the temperature dependence of $\chi$ on field cooling for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$. The SC transition looks broad, which is probably due to the use of powdered samples whose temperature-dependence of $\chi$ is strongly affected by the temperature dependence of the penetration depth. With increasing $p$, the SC transition temperature, $T_c$, increases in the underdoped regime up to $p = 0.153$ and decreases at $p = 0.187$ in the overdoped regime, as shown in Fig. 2(a). The absolute value of $\chi$ at 2 K, $|\chi_{2K}|$, on the other hand, monotonically decreases with decreasing $p$ from $p = 0.187$, as shown in Fig. 2(b).

Here, we discuss the reason for the marked decrease in $|\chi_{2K}|$ with decreasing $p$. First, vortex pinning around twin boundaries is expected to reduce the value of $|\chi_{2K}|$. However, the vortex-pinning effect due to twin boundaries cannot explain the present result, because twin boundaries should appear only for $p \geq 0.124$ where the crystal structure is orthorhombic in $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$. Secondly, the penetration depth increases with decreasing $p$ in the underdoped regime, so that the $|\chi_{2K}|$ value is expected to decrease with decreasing $p$. Assuming the shape of the samples is spherical and referring to the value of the penetration depth at zero temperature of about 200 nm - 500 nm from the optimally doped to underdoped LSCO [16], the typical size of a sample required to explain the result in Fig. 2(b) is calculated to be below 1 $\mu$m in diameter, which is much smaller than that of the present samples, namely, $\sim 40$ $\mu$m. After all, it is reasonable that the $p$ dependence of $|\chi_{2K}|$ is regarded as being due to a decrease

![Figure 1](image_url)

**Figure 1.** Temperature dependence of the magnetic susceptibility, $\chi$, in a magnetic field of 10 Oe on field cooling for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ polycrystals with various $p$ values.
Figure 2. Hole-concentration, $p$, dependences of (a) $T_c$ defined at the cross point between the extrapolated line of the steepest part of the Meissner diamagnetism and zero susceptibility, and (b) the absolute value of $\chi$ at 2 K, $|\chi_{2K}|$, for Y$_{0.8}$Ca$_{0.2}$Ba$_2$Cu$_3$O$_{7-\delta}$ polycrystals. Solid lines are to guide the reader’s eye.

in the SC volume fraction in a SC sample with decreasing $p$.

From the specific heat measurements, on the other hand, it has been found that $\gamma_0$ is not zero but has a finite value even in the SC state for $p = 0.083 - 0.187$. Roughly speaking, moreover, it appears that $\gamma_0$ tends to decrease with increasing $p$ and to be saturated the around optimal doping. These results are quite similar to those observed in LSCO, whereas the values of $\gamma_0$ are obviously larger than those of LSCO [9 - 12]. It is well known in Y-123 cuprates that holes tend to reside in the CuO chain as well as in the CuO$_2$ plane, so that metallic conduction might be realized along the CuO chain [17]. Therefore, the contribution of the CuO chain to the $\gamma_0$ value might be taken into account. Nevertheless, as the $\gamma_0$ value due to the CuO chain is expected to increase or to be almost constant with increasing $p$, the decrease in $\gamma_0$ with increasing $p$ is not simply explained as being due to the contribution of the CuO chain. Moreover, normal carriers in the CuO chain may become SC in the ground state due to the proximity effect from the CuO$_2$ plane. Accordingly, from the results of $\gamma_0$ we anticipate the existence of normal-state carriers in the ground state and probably the existence of non-SC regions in a SC sample. To be conclusive, further study of the contribution of the CuO chain to the $\gamma_0$ value is necessary.

4. Summary
We have investigated the possible phase separation from $\chi$ and specific heat measurements for slightly overdoped and underdoped Y$_{0.8}$Ca$_{0.2}$Ba$_2$Cu$_3$O$_{7-\delta}$ polycrystals. It has been found that, with decreasing $p$, $|\chi_{2K}|$ monotonically decreases suggesting the decrease in the SC volume fraction in a SC sample. Moreover, finite values of $\gamma_0$ have been observed in the ground state for all measured samples, suggesting the existence of normal-state carriers even in the SC state. The present results are quite similar to those observed in LSCO and suggest that a phase separation into SC and normal-state regions takes place in Y$_{0.8}$Ca$_{0.2}$Ba$_2$Cu$_3$O$_{7-\delta}$ as well. It appears that the phase separation is a generic feature in high-$T_c$ cuprates.
References
[1] Pan S H, O’Neal J P, Badzey R L, Chamon C, Ding H, Engelbrecht J R, Wang Z, Eisaki H, Uchida S, Gupta A K, Ng K W, Hudson E W, Lang K M and Davis J C 2001 Nature (London) 413 282.
[2] Loram J W, Mirza K A, Wade J M, Cooper J R, Liang W Y 1994 Physica C 235 134.
[3] Wang Y, Yan J, Shan L, Wen H H, Tanabe Y, Adachi T and Koike Y 2007 Phys. Rev. B 76 064512
[4] Uemura Y J, Keren A, Le L P, Luke G M, Wu W D, Kubo Y, Manako T, Shimakawa Y, Subramanian M, Cobb J L and Merkert J T 1993 Nature (London) 364 605
[5] Niedermayer Ch, Bernhard C, Binninger U, Glückler H, Tallon J L, Ansaldo E J and Budnick J I 1993 Phys. Rev. Lett. 71 1764
[6] Ohsugi S, Kitatoka Y and Asayama K 1997 Physica C 282-287 1373
[7] Tababe Y, Adachi T, Noji T and Koike Y 2005 J. Phys. Soc. Jpn. 74 2893
[8] Tababe Y, Adachi T, Omori K, Sato H, Noji T and Koike Y 2007 J. Phys. Soc. Jpn. 76 113706
[9] Nohara M, Suzuki H, Ishikawa H, Massen Y, Sakai F and Takagi H 2000 J. Phys. Soc. Jpn. 69 1602
[10] Wen H H, Lie Z Y, Zhou F, Xiong J, Ti W, Xiang T, Komiya S, Sun X and Ando Y 2004 Phys. Rev. B 70 214505
[11] Omori K, Adachi T, Tababe Y, Koike Y 2007 Physica C 460-462 1184
[12] Koike Y, Adachi T, Tababe Y, Omori K, Noji T and Sato H 2008 J. Phys. Conf. Series 108 012003
[13] Tranquada J M, Sternlieb B J, Axe J D, Nakamura Y and Uchida S 1995 Nature (London) 375 561
[14] Akoshima M and Koike Y 1998 J. Phys. Soc. Jpn. 67 3653
[15] Obertelli S D, Cooper J R, Tallon J L 1992 Phys. Rev. B 46 14928
[16] Li Q, Sunaga M, Kimura T and Kishio K 1993 Phys. Rev. B 47 2854
[17] Tokura Y, Torrance J B, Huang T C, Nazzal A I 1988 Phys. Rev. B 38 7156