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Correlated Decrease between the Superconducting Volume Fraction and T_c and Possible Phase Separation in the Overdoped Regime of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$

Y. Tanabe, T. Adachi, T. Noji, H. Sato and Y. Koike

*Department of Applied Physics, Graduate School of Engineering, Tohoku University,
6-6-05, Aoba, Aramaki, Aoba-ku, Sendai 980-8579, Japan*

Abstract. In order to prove the possible phase separation in the overdoped high- T_c superconductors, suggested from the muon-spin-relaxation measurements, the superconducting (SC) volume fraction has been investigated in detail in the overdoped regime of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. From measurements of the magnetic susceptibility, χ , using a single crystal in which the Sr concentration, x , continuously changes in the overdoped regime, both the absolute value of χ at 2 K, $|\chi_{2\text{K}}|$, on field cooling reflecting the SC volume fraction and T_c have been found to decrease with increasing x . Moreover, it appears that $|\chi_{2\text{K}}|$ has a roughly linear relation to T_c . It has been concluded that phase separation into SC and normal-state regions takes place in the overdoped regime of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$.

Keywords: magnetic susceptibility, phase separation, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, overdoped regime,

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The inhomogeneity of the superconductivity in the CuO_2 planes is one of the recent central issues in the study of high- T_c superconductivity. Scanning-tunneling-microscopy (STM) experiments in the optimally doped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ have revealed that both the local density of states and the superconducting (SC) gap are spatially inhomogeneous [1]. However, STM is very sensitive to the surface electronic state. Therefore, the inhomogeneity in the CuO_2 plane is still controversial and it is necessary to confirm it from measurements reflecting the bulk nature of a sample.

In the overdoped regime of $\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}$, it has been found from the muon-spin-relaxation (μSR) measurements that the SC carrier density over the effective mass decreases with an increase in the hole concentration, suggesting possible microscopic phase separation into SC and normal-state regions [2]. From measurements of the magnetic susceptibility, χ , in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO), on the other hand, it has been pointed out that the shielding volume fraction estimated from χ at 4.2 K on zero-field cooling using bulk samples is almost 100 % in the range $0.07 \leq x \leq 0.27$ [3]. In general, however, the shielding volume fraction is larger than the real SC volume fraction in the presence of inhomogeneity such as the microscopic phase separation.

In this paper, we have investigated the hole-concentration dependence of the SC volume fraction from the χ measurements on field cooling in the overdoped regime of LSCO [4].

A single-crystal rod with the Sr concentration, x , changing continuously in the overdoped regime was grown by the traveling-solvent floating-zone method. Several pieces of single crystal obtained by slicing the single-crystal rod were used for the χ measurements. The x value of each single-crystal piece was estimated by ICP. The details have been described elsewhere [4]. The χ measurements were carried out at low temperatures down to 2 K, using a superconducting quantum interference device magnetometer.

Figure 1 shows the temperature dependence of χ in a magnetic field of 10 Oe on field cooling for powdered or non-powdered samples of single-crystal LSCO pieces with $x = 0.198 - 0.273$, together with the data of polycrystalline powder samples of LSCO with $x = 0.18, 0.20$ and 0.22 [5]. The SC transition looks broad for the powdered samples, which is ascribed to the temperature dependence of the penetration depth. With increasing x , the absolute value of χ at 2 K, $|\chi_{2\text{K}}|$, as well as T_c decreases and becomes almost zero for $x > 0.273$. As for the data of non-powdered samples of

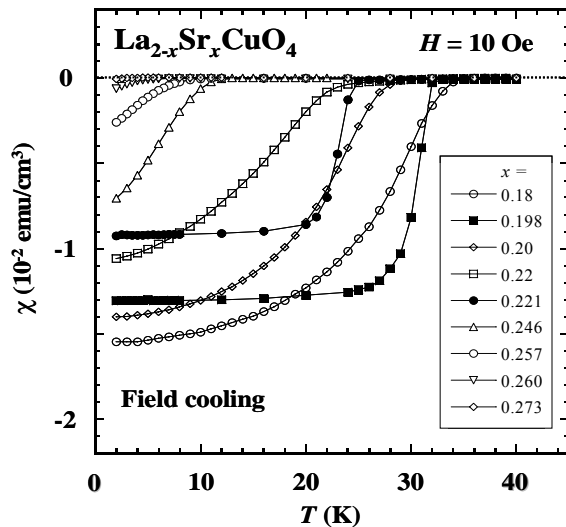


FIGURE 1. Temperature dependence of the magnetic susceptibility, χ , in a magnetic field of 10 Oe on field cooling for single-crystal $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ pieces with $x = 0.198 - 0.273$. Open symbols indicate the data of powdered samples obtained from each single-crystal piece, while closed ones are the data of non-powdered samples of single-crystal pieces in a field parallel to the c axis. The data of polycrystalline powder samples with $x = 0.18, 0.20, 0.22$ are also plotted for reference [5].

$x = 0.198$ and 0.221 , the SC transition is sharp owing to the good quality of the single-crystal pieces, and both T_c and the value of $|\chi_{2K}|$ decrease with increasing x [6]. These results mean the decrease of the SC volume fraction with increasing x in the overdoped regime of LSCO, strongly suggesting that phase separation into SC and normal-state regions takes place.

Figure 2 displays the plot of $|\chi_{2K}|$ vs T_c for powdered samples of single-crystal LSCO pieces. The data of the polycrystalline powder samples of LSCO are also plotted [5]. Both T_c and $|\chi_{2K}|$ seem to decrease cooperatively with increasing x and disappear at $x \sim 0.273$ in the overdoped regime of LSCO. Supposing that the microscopic phase separation suggested from the STM experiment occurred [1], the proximity effect due to the increase of the normal-state regions with increasing x would result in the decrease in T_c .

There are two possible origins of the phase separation in the overdoped regime. One is related to the decrease of the SC condensation energy with increasing x [7]. In this case, phase separation into the hole-poor SC region with $x \sim 0.19$ and the hole-rich normal Fermi-liquid state region with $x \sim 0.30$ will probably take place. The other is due to doping of holes into the $\text{Cu}3d$ orbital, producing free Cu spins

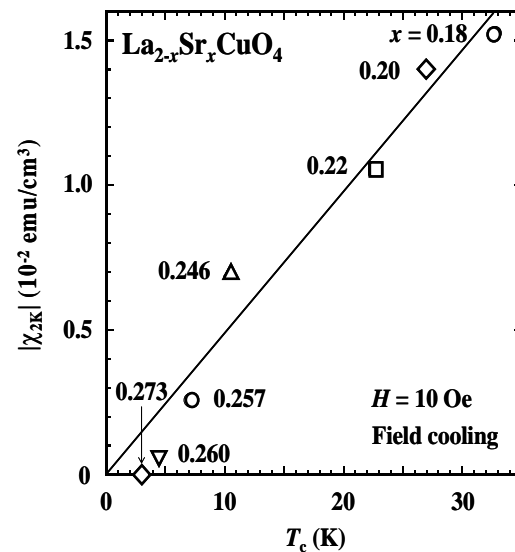


FIGURE 2. $|\chi_{2K}|$ vs T_c for powdered samples of single-crystal LSCO pieces. The data of polycrystalline powder samples with $x = 0.18, 0.20, 0.22$ are also plotted [5].

around the holes [8] and/or disturbing the antiferromagnetic correlation between Cu spins [9]. Both of them will bring about the local destruction of superconductivity around holes, generating normal-state regions in the SC sea.

In summary, it has been found from the χ measurements in the overdoped regime of LSCO that both T_c and $|\chi_{2K}|$ decrease with increasing x , strongly suggesting the phase separation into SC and normal-state regions.

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