ELECTRONIC PHASE DIAGRAM OF La$_{1.875}$Ba$_{0.125-x}$Sr$_x$CuO$_4$

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We performed systematic measurements of magnetic susceptibility on single crystals of La$_{1.875}$Ba$_{0.125-x}$Sr$_x$CuO$_4$. The dependence of the superconducting transition temperature on Sr-concentration demonstrates a step-like pattern upon doping at $x\approx 0.08$ as the crystal structure changes from low-temperature tetragonal (LTT) to low-temperature orthorhombic (LTO) phase at low temperature. Upon cooling, an anomalous upturn in the susceptibility was observed at the structural phase transition between the LTT-LTO phases under the magnetic field parallel to $c$-axis.

1 Introduction

The discovery of anomalous suppression of superconductivity in La$_{2-x}$Ba$_x$CuO$_4$ system with $x\approx 1/8$, an issue dubbed the 1/8 problem, has been paid remarkable attention in the field superconductivity. In this system, a structural phase transition from the low temperature orthorhombic (LTO) to the low temperature tetragonal (LTT) phase occurs in a narrow range of Ba concentrations surrounding 1/8. A similar suppression of superconductivity is also observed in La$_{2-y-z}$Nd$_y$Sr$_z$CuO$_4$ in the LTT phase around $x=1/8$. The superconductivity of La$_{2-x}$Sr$_x$CuO$_4$ in the LTO phase, however, is suppressed around $x=0.115$. Thus, suppression of superconductivity is a generic feature of the hole-doped La-214 system, dependant on crystal structure.

The recent discovery of both spin and charge stripe orders in the LTT phase of the La$_{1.6-x}$Nd$_{0.4}$Sr$_x$CuO$_4$ system reveal a new aspect of the 1/8 problem. A sample with $x\approx 1/8$ possesses the highest stripe ordering temperature and the lowest superconducting transition temperature ($T_c$), suggesting a competition between stripe order and superconductivity. Moreover, since the stripe ordering develops below the LTO-LTT transition temperature ($T_d$), the LTT lattice potential is favorable for the pinning of dynamical fluctuations of spin/charge stripes. The static/quasi-static magnetic order, however, is also observed in orthorhombic La$_{2-x}$Sr$_x$CuO$_4$. These results suggest that stripe order is an inherent property of the La-214 system at $x=1/8$, irrespective of crystal structure, whose stability is affected by the lattice potential.

To clarify the relationship between the suppression of superconductivity, stripe order and crystal structure, we investigated the electronic phase diagram of the La$_{1.875}$Ba$_{0.125-x}$Sr$_x$CuO$_4$ (LBSCO) system. At low temperature, this system allows the crystal structure to change from LTT to LTO through the variation of the Ba/Sr doping ratio while keeping the total hole concentration of 1/8 constant. LBSCO is a suitable compound to investigate the physical properties of the 1/8 problem without the effects of a large, rare-earth moment. In this paper, we measured the magnetic susceptibility of LBSCO single crystals at varying several doping levels. We discovered a clear $x$-dependence of $T_c$ and clarified an anomalous
behavior of the susceptibility.

2 Experimental Details

Single crystals of LBSCO with $x=0.05, 0.06, 0.075, 0.085$ and $0.1$ were grown using a traveling-solvent floating-zone method with two infrared furnaces (Nichiden Machinery, SC-N35HD-E and SC-K15HD) Growth conditions were similar to those used to generate a Ba-free La$_{2-x}$Sr$_x$CuO$_4$ single crystal. Large single crystals with high Ba concentrations are lacking due to difficulties in growth. We have produced single crystal rods with the typical diameter of $\sim 6$mm and a length of $\sim 80$mm by using large, focusing mirrors. Fig.1 exhibits a single crystal rod of La$_{1.875}$Ba$_{0.04}$Sr$_{0.085}$CuO$_4$ $\sim 100$mm in length. A longer crystal rod reduces the concentration gradient in the direction of growth. Magnetic susceptibility measurements on several parts of a crystal rod demonstrate the saturation of $T_c$ at growth lengths greater than $\sim 50$mm. Samples were annealed under oxygen gas flow for 50 hours at $900^\circ$C and then cooled to $500^\circ$C at a rate of $10^\circ$C/h. Following a subsequent annealing at $500^\circ$C for 50 hours, samples were subjected to furnace-cooling to reduced them to room temperature. To determine $T_c$ (onset), we measured the diamagnetic susceptibility at the final part of growth by using a SQUID magnetometer (Quantum Design, MPMS 2 and MPMS XL) under a magnetic field of 10 Oe following the zero-field-cooling process. In a magnetic field of 50000 Oe, parallel to $a_{tetra}$- and c-axes, we measured the temperature dependence of susceptibility for a single crystals of La$_{1.875}$Ba$_{0.075}$Sr$_{0.05}$CuO$_4$.

3 Results and Discussion

We measured the temperature dependence of diamagnetic susceptibility for a series of crystals (Fig.2). Samples with $x=0.05, 0.06, 0.075, 0.085$ and $0.1$ exhibit a superconducting transition at $T_c=10$K, $11.5$K, $14$K, $32$K and $30.5$K, respectively. $T_c$ changes discontinuously, however, upon Sr-doping at a concentration of approx-
Figure 2. Magnetic susceptibility measured at 100e for zero-field-cooled single crystals of La$_{1.875}$Ba$_{0.125-\textit{x}}$Sr$_{\textit{x}}$CuO$_4$ with \textit{x}=0.05, 0.06, 0.075, 0.085 and 0.1.

approximately \textit{x}=0.08; \textit{T}_c is \sim 30K for \textit{x} \geq 0.085 and is \sim 12K for \textit{x} \leq 0.075. The magnetic susceptibilities for \textit{x} \leq 0.075 exhibit an anomaly around 30K, corresponding to the superconducting transition of the residual LTO phase. Such a residual LTO phase is also reported by a recent, high-resolution, neutron power diffraction study examining LBSCO.\textsuperscript{14}

In Fig.3, we summarize the \textit{T}_c of 1/8-doped LBSCO system as a function of Sr concentration \textit{x}. The \textit{T}_c of Ba-free La$_2-x$Sr$_x$CuO$_4$ with either \textit{x}=0.12 or 0.13 are listed as a reference.\textsuperscript{12,13} As \textit{x} increases, \textit{T}_c rises slowly from \textit{x} \leq 0.075, then increases rapidly around \textit{x}=0.08, saturating around \textit{T}=30K for \textit{x} \geq 0.085. By plotting \textit{T}_{d2} as a function of \textit{x} (the dashed line in Fig.3)\textsuperscript{12} a close relation between highly reduced, depending on \textit{T}_{d2} as compared to the LTO phase in which \textit{T}_c is independent of Ba/Sr ratio. This phenomenon is more clearly observed in our present measurements using single crystals compared to previous studies utilizing powder samples.

The graph of \textit{T}_c as a function of \textit{x} reveals a first-order transition-like behavior. In contrast, \textit{T}_{d2} as determined by powder X-ray measurement, demonstrates a gradual change similar to a second-order transition upon doping. A close relation-
Figure 3. Phase diagram of superconducting transition temperatures ($T_c$) for La$_{1.875}$Ba$_{0.125-x}$Sr$_x$CuO$_4$ as a function of $x$ (closed circles) and the LTO-LTT transition temperature ($T_{d2}$) as determined by X-ray diffraction (dashed line). The $T_c$ of Ba-free La$_{2-x}$Sr$_x$CuO$_4$ with either $x=0.12$ (open square) or 0.13 (closed square) is listed. The open circle represents the $T_{d2}$ of a sample possessing an $x=0.05$ as determined by neutron scattering measurement. The solid line demonstrates the overall shape of the $T_{d2}$ function.

ship between crystal structure and suppression of superconductivity should produce a sharp transition in the $x$ dependence of $T_{d2}$ at approximately $x=0.08$. Thus, the $x$-dependence of $T_{d2}$ using single crystals should be examined in more detail.

We measured the magnetic susceptibility of La$_{1.875}$Ba$_{0.075}$Sr$_{0.05}$CuO$_4$ measured under a magnetic field of 50000 Oe parallel to the $a_{tetra}$- or $c$-axes (Fig. 4). Under the latter field, an anomalous upturn is observed at $\sim$38K as the temperature is lowered. This anomalous behavior is independent of the applied field amplitude. The upturn occurs at $T_{d2}$, suggesting a simultaneous change in both the spin and the crystal structure, as discussed with high field MNR experiments on La$_{1.885}$Sr$_{0.115}$CuO$_4$. It is necessary to perform a systematic study of LBSCO to understand the interplay between the anomalous behavior altering magnetic susceptibility, the LTT-LTO transition, and the suppression of superconductivity.
La$_{1.875}$Ba$_{0.075}$Sr$_{0.05}$CuO$_4$

Figure 4. Magnetic susceptibility of La$_{1.875}$Ba$_{0.075}$Sr$_{0.05}$CuO$_4$. Magnetic field of 50000 Oe was applied along the $a_{tetra}$-(closed circles) and c-axes(open circles).

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