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Effect of Composition Disorder in a System of Superconducting Granules on the Superconducting Properties of $La_{2-x}Sr_xCuO_4$ Ceramic Samples

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Abstract—The effect of the degree of doping with strontium has on the superconducting properties of granular superconductor $La_{2-x}Sr_xCuO_4$ is considered. It is found that reducing the concentration of strontium under conditions of compositional disorder broadens the superconducting transition above the temperature of intergranular Josephson coupling and raises the temperature of the completion of the global superconducting transition.

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In this work, we studied features of the resistive superconducting transition of three samples of cuprate $La_{2-x}Sr_xCuO_4$, prepared via standard solid-phase synthesis. Our aim was to study what effect the degree of compositional disorder has on the superconductivity of $La_{2-x}Sr_xCuO_4$ ceramic samples. The strontium content was $x = 0.15$, 0.10, 0.05. Value $x = 0.05$ corresponds to the minimum concentration at which complex oxide $La_{2-x}Sr_xCuO_4$ becomes superconducting. Concentration $x = 0.15$ corresponds to the optimum doping level [1]. All samples were tested at the preparation stage by means of X-ray, magnetic, and electron microscopy. The microstructure, the elemental composition of the samples, and the composition of separate phases were determined via scanning electron microscopy on a Cam Scan scanning electron microscope. The copper and lanthanum content were determined on an EDS LINK AN-10000 spectrometer. The strontium content was determined on a highly sensitive WDS MIKROSPEC spectrometer in five regions of each sample. The ratio of elements in a sample corresponded to its chemical formula. The grain size of the ceramics was ≈1–3 μm.

Ceramic samples of HTSC cuprates with grain sizes of several μm can be described as ensembles of superconducting granules of a type II superconductor. The effect granularity has on the superconducting properties of a ceramic sample of $La_{1.85}Sr_{0.15}CuO_4$ was considered in [2]. It was shown that polycrystalline samples of $La_{1.85}Sr_{0.15}CuO_4$ are heterogeneous systems in which superconducting phase coherence is established at $T < T_{cJ}$ via Josephson coupling between superconducting granules. It was established in [3] that the degree of compositional (and structural) disorder in samples of antiferromagnetic (AFM) $La_{2-x}Sr_xCuO_4$ grows as the strontium content diminishes, leading to smearing of the AFM transition at a sufficient reduction of the impurity concentration. It is thought that in a system of $La_{2-x}Sr_xCuO_4$ superconducting granules, the increase in structural disorder when the degree of doping is lowered can lead to corresponding variations of the superconducting phase transition.

One sign of compositional disorder is structural disorder associated with impurities and vacancies randomly scattered over a crystalline lattice (universal disorder), and with the accumulation of impurities and precipitates of another phase (non-universal disorder) [4]. Let us consider the effect that reducing the level of doping with Sr impurities has on the superconducting phase transition.

The curves of the temperature dependence of magnetic susceptibility for the three samples under study were obtained using a SQUID MPMS-XL5 magnetometer. Temperatures T_{c01} of the superconducting transition, determined from the diamagnetic response signal (Fig. 1), are given in Table 1. Temperature T_{c01} falls along with the content of strontium. Superconductivity is suppressed at $H = 1$ T for the sample with the minimum concentration of strontium ($x = 0.05$). A diamagnetic response for this sample was detected only at weak field $H = 10$ Oe.

The temperature dependences of the resistivity of samples 1 and 2 demonstrate the two-step character of the transition (Fig. 2) typical of heterogeneous superconductors. Transitions of this kind were considered

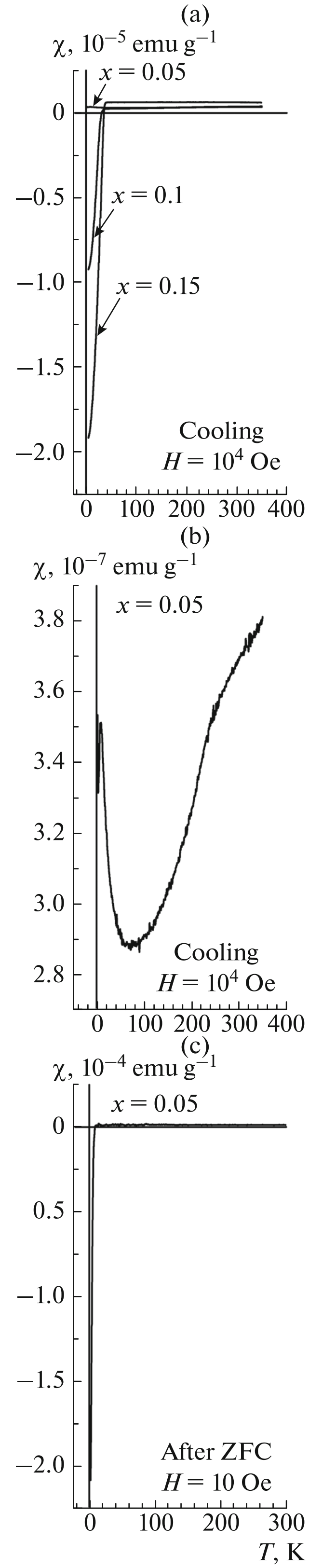

Fig. 1. Temperature dependences of the magnetic susceptibility of $La_{2-x}Sr_xCuO_4$: (a) cooling in a field of 10^4 Oe; (b) enlarged scale of the $\chi(T)$ dependence for $x = 0.05$; (c) $\chi(T)$ dependence for $x = 0.05$, recorded in the heating regime at $H = 10$ Oe after cooling in a zero field.

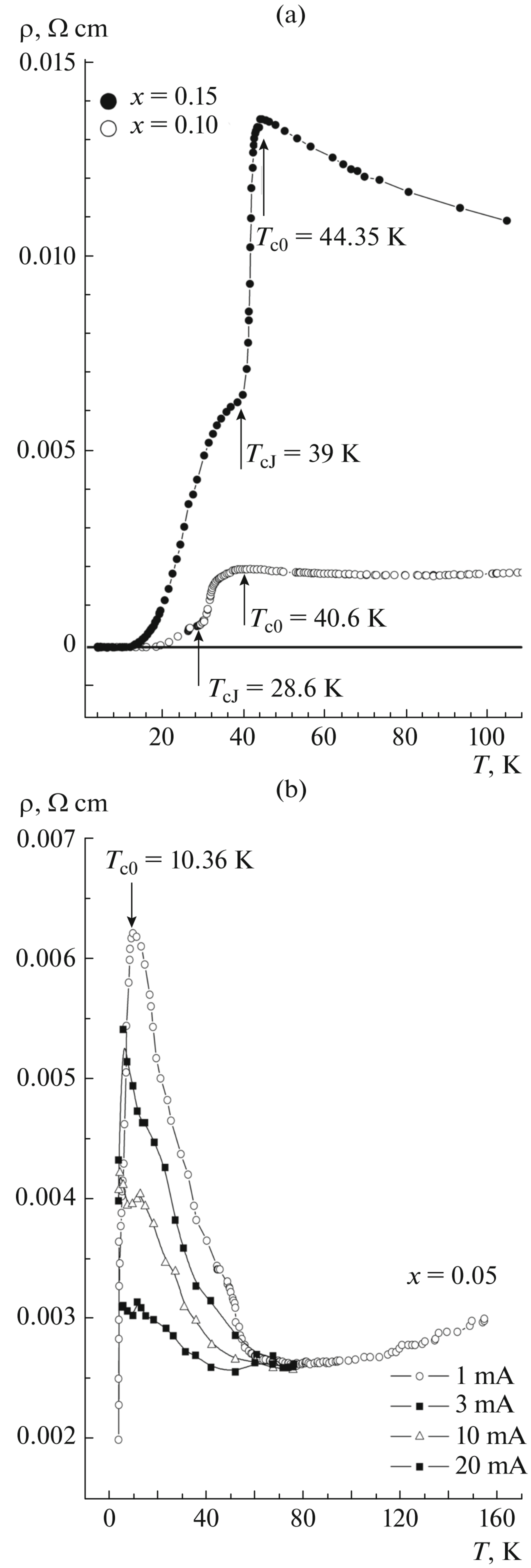

Fig. 2. Temperature dependences of the resistivity of $La_{2-x}Sr_xCuO_4$ samples: (a) superconducting transitions of $La_{1.85}Sr_{0.15}CuO_4$ and $La_{1.9}Sr_{0.1}CuO_4$ samples, recorded at a current of 3.0 mA; (b) temperature dependences of the resistivity of $La_{1.95}Sr_{0.05}CuO_4$ sample, recorded at different currents.

Table 1

Sample	x	χ, emu g^{-1} $H = 10^4$ Oe, $T = 5$ K	χ, emu g^{-1} $H = 10$ Oe, $T = 5$ K	T_{c01}, K $H = 10^4$ Oe	T_{c01}, K $H = 10$ Oe
1	0.15	-1.9×10^{-5}	–	36.0	39.0
2	0.10	-9.2×10^{-6}	-0.0013	31.5	36.2
3	0.05	3.3×10^{-7}	-6.2×10^{-5}	–	8.6

Table 2. T_c is the superconducting transition temperature: $\Delta T_1 = T_{c0} - T_{cJ}$; $\Delta T_2 = T_{cJ} - T_c$

J, μA	T_{c0}, K	T_{cJ}, K	T_c, K	ΔT_1, K	ΔT_2, K	T_{c0}, K	T_{cJ}, K	T_c, K	ΔT_1, K	ΔT_2, K
	Sample 1					Sample 2				
100	46.8	40	21	6.8	19	43.6	32	22	11.6	10
300	45.5	39	20	6.5	19	43.6	30	20.6	13.6	9.4
3000	44.35	39	12	5.35	27	40.6	28.6	16.2	12	12.4
10000	36	31	—	5	—	32	20	4.4*	12	15.6

* $T_c = 4.4$ K is the nominal temperature of the superconducting transition (see text).

in detail in [5]. Temperature T_{c0}, obtained under the condition $d\rho/dT = 0$, corresponds to the transition of separate granules to the superconducting state. The nonmetallic behavior of $\rho(T)$ at $T > T_{c0}$ ($d\rho/dT < 0$) is a consequence of weak intergranular coupling; i.e., $E_J \ll T_{c0}$, where E_J is the Josephson energy. Temperature T_{cJ} corresponds to the system transitioning to the global superconductivity state ($E_J \gg T_{cJ}$) when phase coherence between separate granules is established. The superconducting transition in a macroscopic system is complete when the granules, strongly interacting with one another, form an infinite superconducting cluster [2]. Temperatures T_c of the superconducting transition's completion, corresponding to different measuring currents, are indicated in Table 2. The resistive superconducting transition for sample 3 was not detected at currents $J \geq 1$ mA. However, at minimum current $J = 1$ mA, we observe in Fig. 2b the onset of the superconducting transition at $T_{c0} = 10.36$ K.

Comparison of the results presented in Tables 1 and 2 for T_{c01} and T_c measurements shows that the diamagnetic response is observed only at the instant a sufficiently large volume of the superconducting phase forms, and if there is phase coherence between superconducting granules.

Dependences $\rho(T)$, measured at different values of the measuring current (in the range of 10 μA–10 mA for $x = 0.15$ and 100 μA–40 mA for $x = 0.10$), demonstrated the rather strong dependence of the resistive superconducting transition on J. As was shown in [2], an increase in current at concentration $x = 0.15$ results in significant broadening of the superconducting transition in the region of $T < T_{cJ}$, starting from critical value $J_c = 3$ mA upon a slight reduction in T_{c0} (by ≈ 0.5 K). When the current is raised further, some of the weakest links in the percolation chains with the least critical current change to a resistive state, increasing the general resistivity of the system, and temperature T_{c0} of the transition falls substantially at $J = 10$ mA. Similar variation in the nature of the transition with increasing J was observed for sample 2 with concentration of strontium $x = 0.10$ (see Table 2). It should be noted that $T_c = 4.4$ K at $J = 10$ mA (Table 2) should be considered nominal for this sample, since the superconducting transition is not yet complete at this minimum temperature. A comparatively weak electric current thus initially destroys the phase coherence between separate granules, as a result of which the resistivity grows sharply at $J = 3$ mA in the region of $T < T_{cJ}$, and the superconducting transition is substantially broadened (ΔT_2 increases). Upon a further increase in current, the intragranular superconductivity is destroyed and temperature T_{c0} of the superconducting transition is reduced. A similar effect has a magnetic field (Fig. 3). It follows from the data presented in Figs. 2, 3, and Table 2 that the critical fields and currents for superconducting granules considerably exceed those for an intergranular medium.

A rather sharp superconducting transition for sample 1 ($x = 0.15$) occurs at $H = 0.005$ T with a small shoulder below T_{cJ} (Fig. 3a). The broadening of the transition at $T \leq T_{cJ}$ was observed in a field of 0.01 T, and only in a field of $H > 1$ T was temperature T_{c0} appreciably reduced (by ≈3.6 K at $H = 14$ T). The broadening of the transition at $T \leq T_{cJ}$ for sample 2 ($x = 0.10$) was observed in lower fields of $H = 0.002$ T,

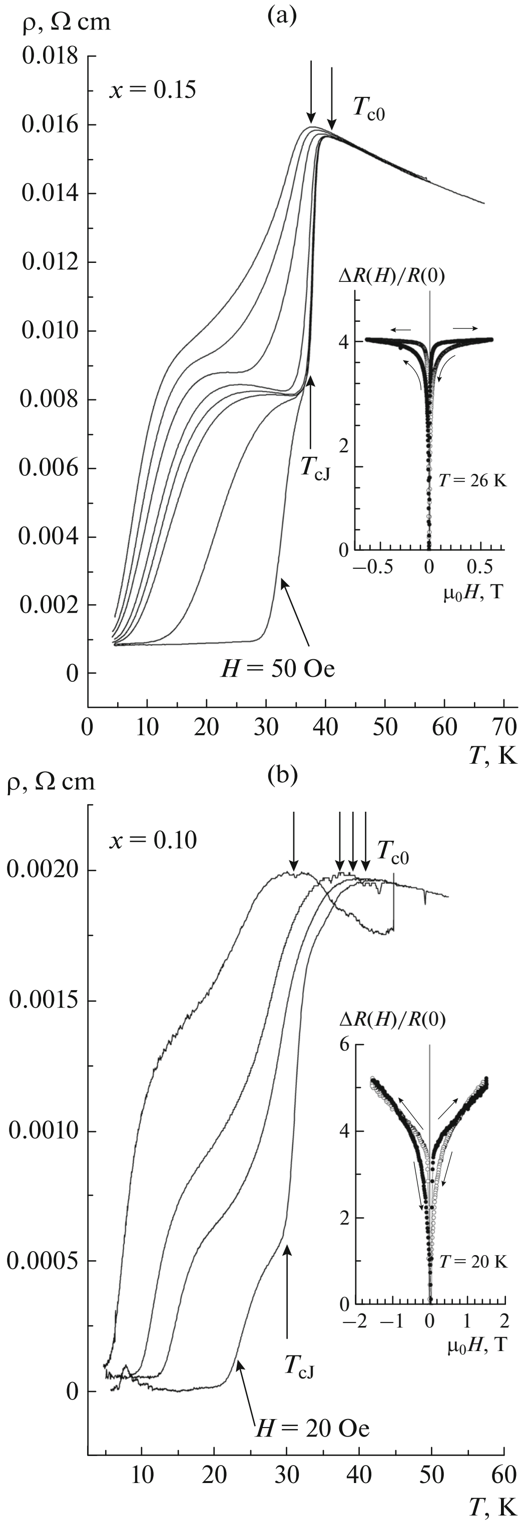

Fig. 3. Temperature dependences of the resistivity of $La_{2-x}Sr_xCuO_4$ samples, recorded in different magnetic fields $\vec{H} \perp \vec{J}$: (a) $H = 50$, 100, 800, 2×10^3, 10^4, 5×10^4, 10^5, 1.4×10^5 Oe; (b) $H = 20$, 2×10^4, 5×10^4, 1.4×10^5 Oe. Magnetoresistive effects recorded at $T < T_{cJ}$ and currents (a) $J = 1.5$ mA and (b) $J = 150$ μA are shown in the inserts in Figs. 3a, 3b.

and T_{c0} fell much more upon an increase in the field than for sample 1 (by ≈9 K in a field of 14 T).

The magnetoresistive effect in the inserts in Figs. 3a, 3b corresponds to the behavior of a heterogeneous system, described on the basis of the two-level model of a critical state in [6]. There is a clockwise hysteresis.

According to the data of Table 2, Fig. 2a, and Fig. 3, variations in the superconducting parameters are observed when the concentration of strontium is reduced:

(1) At all values of J, reducing the concentration of strontium results in stronger smearing of the transition above T_{cJ}: ΔT_1 ($x = 0.15$) < ΔT_1 ($x = 0.10$), due possibly to the dependence of E_J on distance d between superconducting granules ($E_J \propto d^{-1}$), which inevitably grows when the concentration of strontium is reduced.

(2) The width of the transition shrinks along with concentration x when Josephson coupling is established between superconducting granules ($T < T_{cJ}$).

(3) Reducing the concentration of strontium raises temperature of transition T_c.

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REFERENCES

1. Masayuki, I., *J. Phys. Chem. Solids*, 2004, vol. 65, p. 1381.
2. Dalakova, N.V., Elekoeva, K.M., Kashezhev, A.Z., Manukyants, A.R., Ponegev, M.Kh., Prokhorenko, A.D., and Sozaev, V.A., *Bull. Russ. Acad. Sci.: Phys.*, 2014, vol. 78, no. 4, p. 337.
3. Dalakova, N.V., Belevtsev, B.I., Belyaev, E.Yu., et al., *Low Temp. Phys.*, 2014, vol. 40, p. 397.
4. Varlamov, A.A. and Larkin, A.I., *Teoriya fluktuatsii v sverkhprovodnikakh* (Fluctuation Theory in Superconductors), Moscow: Dobrosvet, 2007.
5. Dmitrenko, I.M., *Fiz. Nizk. Temp.*, 1986, vol. 14, p. 1045.
6. Ji, L., Rzchowski, M.S., Anand, N., and Tinkham, M., *Phys. Rev. B*, 1993, vol. 47, p. 470.

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