Same Superconducting Criticality for Underdoped and Overdoped $La_{2-x}Sr_xCuO_4$ Single Crystals

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By measuring the superconducting diamagnetic moments for an underdoped and an overdoped $La_{2-x}Sr_xCuO_4$ single crystal with equal quality and roughly equal transition temperatures, it is found that the underdoped sample has only one transition which corresponds to H_{c2}, but the overdoped sample has two transitions with the higher one at H_{c2}. Further investigation reveals the same upper-critical field H_{c2} for both samples although the overall charge densities are very different, indicating the possibility of a very direct and detailed equivalence of the superconducting condensation process in the two doping limits. The second transition for the overdoped sample can be understood as the bulk coupling between the superconducting clusters produced by macroscopic phase separation.

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The mechanism of high temperature superconductors (HTS), one of the challenging issues, has stimulated enormous effort in recent years. Connected with it is a widely accepted electronic phase diagram which contains three major phases: hole underdoped, optimally doped, and overdoped. The contrasting properties in the normal state between an underdoped and an overdoped sample tempt to ascribe the superconductivity to different condensation processes and thus different criticalities. One example is the recently proposed model of considering the superconducting transition of HTS as a Bose-Einstein condensation in the underdoped region and the BCS-like origin in the overdoped region [1,2]. Therefore, it remains unclear whether the HTS has the same condensation process when going through from the underdoped region to the overdoped one. Another puzzling point in an overdoped HTS is that the transition temperature T_c drops with an increase of the number of charge carriers (here the doped holes), in sharp contrast with what appears in the underdoped region. The crossover from the non-Fermi liquid behavior in the underdoped region to the Fermi liquid behavior in the overdoped region with increasing doping level clearly indicates that most of the doped holes participate in the conduction in the normal state. Recent data from the measurement on the penetration depth λ [3,4] show that, however, the superfluid density ρ_s behaves just like the transition temperature T_c, i.e., decreases with the doped hole number. The consequence is that in the overdoped region, the more charge carriers are doped, the lower the superfluid density ρ_s will be. Therefore, the doped holes in the overdoped region seem to be separated into two parts; only part of them condense into a lower energy state leading to the superconductivity. In our previous paper [5] it was shown that the macroscopic phase separation may have occurred in overdoped $Bi_2Sr_{2-x}La_xCuO_{6+y}$ single crystals although we were not sure whether this phase separation is induced by the inhomogeneity of excess oxygen or by the electronic instability. In this Letter, we present a comparative investigation on an underdoped and an overdoped single crystal. A similar two-step transition has been observed only in the overdoped sample leading to an intuitive inference that the macroscopic phase separation may have occurred in the overdoped sample due to the electronic instability rather than the chemical inhomogeneity since in the present system the incorporation of excess oxygen is very difficult especially for the overdoped samples.

Single crystals measured for this work were prepared by the traveling solvent floating-zone technique [6]. A series of single crystals has been investigated for this study. For the sake of simplicity, in this Letter we present the measurement only for two typical single crystals, one underdoped and another one overdoped with almost the same transition temperatures and equal qualities. Figure 1 shows the superconducting transitions of these two typical samples with dimensions of about 2 mm(length) $\times$ 1 mm(width) $\times$ 0.3 mm(thickness) measured with a superconducting quantum interference device (SQUID, Quantum Design, MPMS 5.5). Resistive measurements on these samples show very narrow transition widths (<1 K) indicating a high quality of the samples. The transition temperatures of the overdoped ($x = 0.24$) and the underdoped ($x = 0.092$) samples are 25 and 26 K, respectively, which fall exactly onto the general parabolic curve of T_c versus doping level with optimal doping at about 0.16 and $T_c = 38.5$ K as found by many others [6,7]. The roughly identical qualities and transition temperatures between the underdoped and the overdoped samples provide us an effective way to do the comparative investigation.

Distinct diamagnetic behaviors have been found and shown in Fig. 2 for both samples when a relatively strong external magnetic field is applied. It is clear that for the underdoped sample, there is only one transition marked here as T_{c1}. The slight diamagnetic moment appearing above T_{c1} is due to the fluctuation effect. For the overdoped

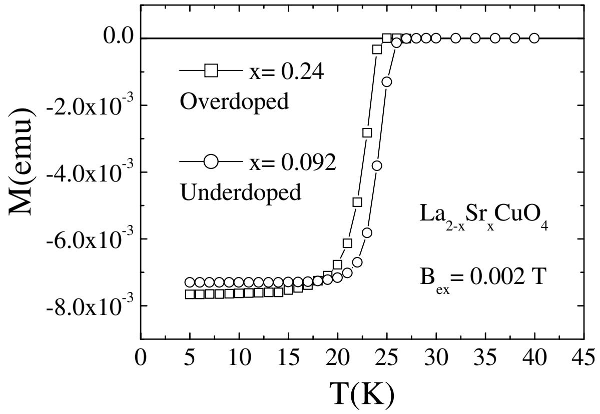

FIG. 1. Temperature dependence of the diamagnetic moments for two typical $La_{2-x}Sr_xCuO_4$ single crystals with $x = 0.092$ (underdoped) and 0.24 (overdoped) measured under an external field of 0.002 T in the zero-field-cooling (ZFC) process. In the ZFC process, the sample is first cooled to a desired temperature at zero field and then an external field is applied; the data are collected in the warming up process with field. It is clear that the underdoped sample and the overdoped sample have equal quality and roughly equal transition temperatures, leading to an effective comparison between these two extreme situations.

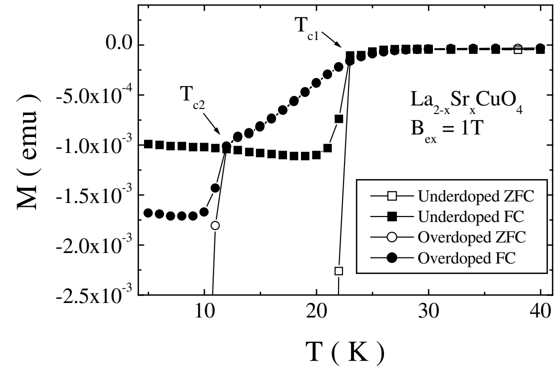

FIG. 2. Temperature dependence of the diamagnetic moments measured for the underdoped sample and the overdoped sample at an external field of 1 T in the ZFC and FC processes. In the FC process, the sample is first cooled to the desired temperature under a field and the data are collected in the warming up process with field. It is evident that there is only one transition for the underdoped sample but two transitions for the overdoped sample. The first transition at T_{c1} for the overdoped sample coincides with the solitary transition of the underdoped sample. This transition shifts slowly with external field, in sharp contrast to the quickly moved second transition at T_{c2} for the overdoped sample.

sample, however, there are two transitions; one appears at almost the same temperature as the underdoped sample, i.e., T_{c1}, while another sharp transition occurs at T_{c2}. The irreversibility for flux motion appears immediately after T_{c1} for the underdoped sample and after T_{c2} for the overdoped one. The behavior of two transitions on one single $M(T)$ curve was previously found in $Bi_2Sr_{2-x}La_xCuO_{6+y}$ single crystals [5] in which the excess oxygen may be inhomogeneous and thus the first transition was attributed to the appearance of superconductivity on some individual clusters (with less oxygen and/or holes) and the second transition is due to the Josephson coupling or proximity effect between these clusters. The two transitions in our present overdoped sample can get the same explanation but clearly the superconducting clusters here are not formed by the inhomogeneity of excess oxygen, rather by the electronic phase separation effect on the holes.

Although the underdoped and the overdoped samples investigated here have almost the same superconducting transition temperatures at zero field, it gives, however, no reason to believe that the two samples have the same criticality at a high magnetic field since they have very different overall hole densities. The superconducting criticality, such as the upper critical field $B_{c2}(T)$, contains important information about the superconducting condensation and probably is also related to the pairing mechanism of Cooper pairs; therefore it is interesting to investigate the criticality of the underdoped and the overdoped samples. For this purpose, we determined the upper critical field $B_{c2}(T)$ for both samples. For the underdoped sample, this is quite easy since there is only one sharp transition. For the overdoped sample, the reversible region is wide and the transition near T_{c1} is rounded; therefore one should use the critical fluctuation theory [8] to derive $B_{c2}(T)$. In Fig. 3, the temperature dependence of the reversible magnetic moments measured under six magnetic fields (0.2 to 5 T) for the overdoped sample are shown. There is a common crossing point at (T^*, M^*) on these curves suggesting strongly an underlying scaling behavior. According to the fluctuation theory of Ullah and Dorsey [8], a general scaling law for high temperature superconductors reads

$$\frac{M}{(B_{\text{ex}}T)^{\alpha}} \propto G\left[\frac{T - T_c(B_{\text{ex}})}{(B_{\text{ex}}T)^{\alpha}}\right], \qquad (1)$$

where $G(x)$ is an unknown scaling function, $\alpha = 2/3$ for 3D and $1/2$ for 2D, M is the magnetic moment, and B_{ex} is the external field. The information about the upper critical field is included in the relation $T_c(B_{\text{ex}})$ or, vice versa, $B_{c2}(T)$; i.e., a correct choice for the relation $T_c(B_{\text{ex}})$ will collapse all the $M(T)$ curves onto one master line. Above scaling law has been well checked for various HTSs [9] delivering a high slope of $B_{c2}(T)$ near T_c. As shown by the inset in Fig. 3, by assuming $B_{c2}(T) = (T - T_c) \times (dB_{c2}/dT)$, a good scaling can be obtained by taking $\alpha = 2/3$ (3D) and $dB_{c2}/dT = -0.7 \pm 0.3$ T/K. The $B_{c2}(T)$ for the overdoped sample determined by doing above scaling and that for the underdoped sample determined directly from the sharp transition at T_{c1} are plotted together in Fig. 4. It is remarkable that both curves are very close to each other. *This is our central result which indicates the same superconducting criticality for the underdoped and the overdoped samples.* It is important to note that for the underdoped sample, the correct way to determine B_{c2} is also to do the critical scaling. Since in

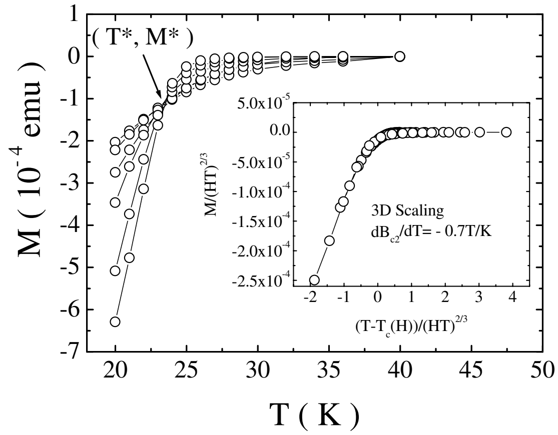

FIG. 3. Temperature dependence of the reversible magnetic moments for the overdoped sample at fields of 0.2, 0.4, 1.0, 2.0, 3.5, and 5 T. A clear common crossing point appears at (T^*, M^*) strongly suggesting an underlying scaling behavior. The inset shows the scaling of the data according to Eq. (1) by taking $T_c = 25$ K, $\alpha = 2/3$, and $dB_{c2}/dT = -0.7$ T/K.

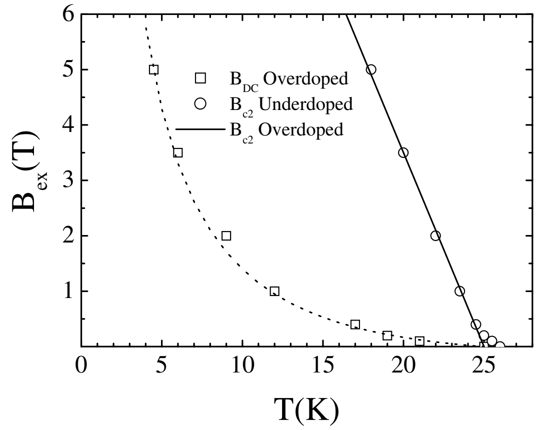

FIG. 4. The upper critical field B_{c2} determined for the underdoped sample (open circles) directly from the sharp transition at T_{c1}, and for the overdoped sample (solid line) from the scaling shown in the inset in Fig. 3. The upper critical fields for these two samples are very close to each other indicating the same criticality for superconductivity in these two samples albeit the overall hole densities are very different. The open squares represent the second transition of the overdoped sample and the dotted line is a guide to the eyes.

our present sample the fluctuation region is too small to do that, therefore, we determined the B_{c2} directly from the sharp transition at T_{c1}. For underdoped $YBa_2Cu_3O_{7-\delta}$, for example [9], the fluctuation region is wide and then the transition is not sharp, one should use the critical fluctuation theory to determine $B_{c2}(T)$.

Now we turn to the second transition at T_{c2} on the $M(T)$ curve for the overdoped sample. As shown by the open squares in Fig. 4, the transition line at T_{c2} is extremely positive curved, being very similar to the so-called $H_{c2}(T)$ line determined from the resistive measurement by Mackenzie *et al.* [10] in overdoped Tl-2201 samples. As argued in our previous paper [5], this transition is not corresponding to the upper critical field $B_{c2}(T)$ but corresponds rather to the Josephson coupling [11] or proximity effect between the superconducting clusters preformed at T_{c1}.

In order to explain the data, we have proposed the following picture: in the overdoped region, some superconducting clusters can be formed via electronic phase separation. These clusters are surrounded by the good metallic regions with rich holes. By lowering temperature, these clusters become superconductive first at T_{c1} and the bulk superconductivity is established probably via Josephson coupling or the proximity effect between these clusters at a lower temperature T_{c2}. One may argue that the two-step transition for our present overdoped sample is induced by some extrinsic causes, for example, the possible presence of the second chemical phase. This can, however, be ruled out by the observation of very clean $(00l)$ peaks from the x-ray diffraction (XRD) pattern, and the symmetric nonsplitting Laue spots on the present overdoped sample. This argument stands also weakly against the same magnitude of the FC diamagnetic moments after T_{c1} and T_{c2} as shown in Fig. 2 if they would have been due to two chemical phases, then they should have been distinguished by XRD. Another possible argument may be that there is a nearby first order (orthorhombic to tetragonal) phase transition at around $x = 0.20$ as argued in the past by Takagi *et al.* [2], which probably leads to an intrinsic chemical inhomogeneity. The two-step transition observed in our overdoped sample is certainly not induced by this possible phase transition because of the following reasons: (i) This two-step behavior has been observed both below and above $x = 0.20$ in the overdoped region, (ii) it has no reason to believe that one of the chemical phases (if it exists) should have the same criticality as the corresponding underdoped sample, and (iii) the two-step behavior has been observed in many different families of overdoped samples, for some of them without orthorhombic to tetragonal phase transition.

The picture derived from our measurement inhibits taking the overdoped sample as a system with uniformly distributed fermions and thus refuses the new theories based on this consideration. As argued by Kivelson and Emery [13], in a system with a local tendency to phase separation, one has some kind of "Coulomb-frustrated phase separation"; i.e., the system is inhomogeneous on an intermediate scale. Since these phase separated clusters are small, the proximity effect should be operational. In this scenario, as the system gets more overdoped, the fraction of the superconducting part shrinks and probably the typical size of the "superconducting clusters" decreases, so the T_c will be suppressed by the proximity effect. This naturally explains the decrease of T_c with doping level and the second step on the $M(T)$ curve in the overdoped region. According to our picture, the more holes are doped, the easier it is to observe the second transition. In regard of inhomogeneity and/or phase separation, our picture can get support from substantial recent experiments done on overdoped samples, such as overdoped Tl-2201 [10,14,15], $Bi_2Sr_{2-x}La_xCuO_{6+y}$ [5], $La_{2-x}Sr_xCuO_{4+\delta}$ [16,17], and $(Y_{1-x}Ca_x)Ba_2Cu_3O_{7-\delta}$ [18,19]. Radcliffe *et al.* [14] measured the electronic specific heat of overdoped Tl-2201 single crystals and found that the $B_{c2}(T)$ determined from the specific heat measurement is much higher than that determined from the magnetoresistance measurement. This conclusion is consistent with our picture. A similar anomaly of B_{c2} was observed by Blumberg *et al.* [15] in the measurement of electronic Raman scattering on overdoped Tl-2201 samples in a high magnetic field. Tallon *et al.* [16] reviewed the muon-spin-rotation measurement on overdoped Tl-2201 and $La_{2-x}Sr_xCuO_{4+\delta}$ and claimed the coexistence of two different regions with different superconducting properties arising from the phase separation. A similar result was also obtained by Ohsugi *et al.* [17] in the nuclear-magnetic-resonance (NMR) measurement in $La_{2-x}Sr_xCuO_{4+\delta}$. Another indirect evidence for the phase separation in the overdoped region was from the extended x-ray absorption fine structure measurement on $(Y_{1-x}Ca_x)Ba_2Cu_3O_{7-\delta}$ samples by Kaldis *et al.* [18] who concluded that the structure of overdoped $(Y_{1-x}Ca_x)Ba_2Cu_3O_{7-\delta}$ samples may be a martensitic form of the optimum doped crystals. This may provide a reasonable explanation to the two energy gaps found in overdoped $(Y_{1-x}Ca_x)Ba_2Cu_3O_{7-\delta}$ from the time-domain spectroscopic measurement [19]. A direct confirmation to our picture would, however, come from the scanning-tunneling-microscopic measurement at different temperatures under a magnetic field. It can carry out the information of the spatial resolved single-electron tunneling spectrum provided that the clusters are static after phase separation and thus deserves certainly further investigation. Our picture may have two folds of impact on theoretical development: First, in HTS there may be only one pairing mechanism which should get a full reflection in the underdoped region, e.g., the pseudogap [20,21] and stripe phase [13], etc. Second, any theory for mechanism of HTS should cover an explanation to the macroscopic electronic phase separation in the overdoped region.

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