Intrinsic percolative superconductivity in heavily overdoped high-temperature superconductors

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Abstract. – Magnetic measurements on heavily overdoped La2−xSrxCuO4, Tl2Ba2CuO6, Bi2Sr2CuO6, Y1−x(Cax)Ba2Cu3O7−δ and Bi2Sr2CaCu2O8 single crystals reveal a new type of magnetization hysteresis loops characterized by the vanishing of the usual central peak near zero field. Since this effect has been observed in various systems with very different structural details, it probably reflects a generic behavior for all high-temperature superconductors. This easy penetration of magnetic flux can be understood in the picture of percolative superconductivity due to the inhomogeneous electronic state in heavily overdoped regime.

The mechanism of high-temperature superconductors (HTS) remains puzzling and one of the most attractive topics in condensed-matter physics. An essential focus lies on the understanding of the parabolically shaped superconducting area in the generic electronic phase diagram containing several phases depending on the hole doping level: underdoped, optimally doped and overdoped. The contrasting properties in the normal state between an underdoped and an overdoped sample tempt us to ascribe the superconductivity to different condensation processes. For example, one picture suggests that the superconducting transition is a Bose-Einstein condensation in the underdoped region and a BCS-type condensation in the overdoped region [1, 2]. One of the problems encountered in an overdoped HTS is that the transition temperature $T_c$ drops with increasing the number of charge carriers (here the doped holes), in sharp contrast with what happens in the underdoped region. The crossover from the non-Fermi liquid in the underdoped region to the approximate Fermi-liquid behavior in the overdoped region may indicate that most of the doped holes join the conduction in the normal state. Recent data from the measurement on the penetration depth $\lambda$ [3] show that, however, the superfluid density $\rho_s$ drops with the doping level $p$ when $p > 0.19$. This leads to a difficult point: in the heavily overdoped regime, the more charge carriers are doped, the less

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the superfluid density $\rho_s$ will be. Therefore, the doped holes in the overdoped region seem to be separated into two parts, and only part of them will condense into a lower-energy state leading to superconductivity.

In our previous papers \[4\], we have presented a preliminary evidence to show that the electronic state in the overdoped region may be inhomogeneous \[5\]. According to this picture, superconducting condensation will occur first in some local region (with less holes) to form some tiny superconducting islands. The extra holes on these superconducting islands will be expelled to the surrounding area to form a hole-rich non-superconducting metallic sea and the bulk superconductivity will be established by the Josephson coupling or proximity effect between these islands. We attributed this inhomogeneity to the “macroscopic” electronic phase separation since a two-step diamagnetic behavior was observed \[4\], one at the upper boundary for superconductivity $H_{c2}(T)$ and another one at much lower values of $T$ and $H$. The term “macroscopic” here means only that the size of superconducting region after the phase separation is at least larger than the coherence length $\xi$ leading to the phase coherence. A similar picture about phase separation in the overdoped region called “Swiss-Cheese” model was proposed early by Uemura \[6\]; the Swiss-Cheese model involves a microscopic inhomogeneity. By measuring and scaling the residual resistivity of the Zn-substituted high-$T_c$ superconductors, Fukuzumi et al. \[7\] suggested that the overdoped regime could be inhomogeneous and the residual resistivity scales with $1 - p$ instead of the hole number $p$. Therefore it always warrants further investigations until it is finalized with solid experimental evidence. If the picture of inhomogeneity is correct, one should be able to observe the percolative superconductivity in the heavily overdoped region. In this letter, for the first time, we present a new type of magnetization hysteresis loop (MHL) characterized by the vanishing of the central penetration peak. This effect can be explained quite well by the picture based on the percolative superconductivity.

Single crystals measured for this work were prepared by the traveling solvent floating-zone technique (LSCO, Bi-2201) and the self-flux method using CuO as flux (TI-2201, Bi-2212 and Y(Ca)-123). Several single crystals have been investigated for this study. Our major conclusion here has been re-checked by using many crystals of LSCO, TI-2201, Bi-2201, Bi-2212 and Y(Ca)-123. The data from Bi-2201, Y(Ca)-123 and Bi-2212 will not be shown here due to the length limit of this paper. All these single crystals have rather narrow superconducting transitions as observed from the temperature-dependent magnetization measurement. The X-ray diffraction pattern (XRD) taken from these single crystals show very good crystallinity without any trace of a second phase. All these guarantee the generality of the major conclusion drawn in this paper. A Quantum Design superconducting quantum interference device (SQUID, MPMS 5.5 T) and an Oxford vibrating sample magnetometer (VSM 3001, 8 T) were used to measure the MHL.

For a uniform superconductor in the superconducting state, the condensate of the superconducting electrons will expel the external field. When the external field $H$ is higher than the lower critical field $H_{c1}$, many quantized magnetic vortices will be formed and penetrate into the interior of the sample. The spatial distribution of the density of these vortices, also called flux profile, is illustrated in the inset of fig. 1. The curve of $M$ vs. $H$ will deviate from the linear relation $M = -H/4\pi$ (Meissner state) at $H_{c1}$ for a perfect cylinder. The magnetization $M$ will continue to grow until the flux front meets at the center of the sample ($B_e = B_p$). By further increasing the external field, more and more magnetic vortices will creep into the sample, and the magnetization will start to drop. Therefore, for any uniform superconductor, a penetration peak near the zero field will appear due to the non-easy penetration of magnetic flux. As an example, in fig. 1 we show one MHL for a YBa$_2$Cu$_3$O$_{7-\delta}$ single crystal near optimal doping. A clear penetration peak can be observed here. When the field drops
Fig. 1 – A typical MHL measured for a YBa$_{2}$Cu$_{3}$O$_{7-\delta}$ single crystal at $T = 30$ K. A clear penetration peak appears near the zero field. This central penetration peak remains until $T \geq T_c$. The inset shows the magnetic-flux profile in a superconducting cylinder at different fields. When $B_e = B_p$, the magnetization reaches the maximum.

down from a high value (from both the positive and the negative side), a huge magnetization peak will appear near the zero field due to the establishing of a large superconducting current. This huge central peak envelops the penetration peak. One can imagine that the penetration peak and the central peak may disappear for a percolative superconducting system due to the difficulty of establishing a high current density near the zero field.

As the magnetic flux fully penetrates into the sample, some vortex phases can be formed in some specific temperature and field region. For example, a picture based on the crossover from the Bragg glass in the low-field region to the vortex glass in the high-field region [8] was proposed to interpret the second-peak effect [9] (narrow and sharp but almost temperature independent) in Bi-2212 single crystals as shown in fig. 2. For YBa$_{2}$Cu$_{3}$O$_{7-\delta}$ single crystals, a broad and temperature-dependent second peak [10] has been observed which has received

Fig. 2 – A typical MHL measured for a Bi$_2$Sr$_2$CaCu$_2$O$_8$ single crystal at $T = 25$ K. A clear penetration peak can be observed near the zero field. This central penetration peak remains until $T \geq T_c$. At a higher field, a second peak appears which is explained as the crossover from the low-field Bragg glass to the high-field vortex glass.
Fig. 3 – MHLs measured for (a) an overdoped \((x = 0.24, T_c = 25 \text{ K})\) and (b) an underdoped \((x = 0.096, T_c = 26 \text{ K})\) LSCO single crystals. For the underdoped sample, a clear central penetration peak is observed, while for the overdoped sample the central peak is too small to be observable, showing an easy-penetration process.

For a heavily overdoped HTS, the situation could be different according to the picture of electronic inhomogeneity. Here we have intentionally investigated the magnetic-flux penetration process for such samples. In fig. 3(a), a typical MHL measured at 9 K for an overdoped LSCO \((x = 0.24)\) is shown. Clearly the central peak becomes too small to be observable. From an enlarged view shown in the inset to fig. 3(a) one can still see a very tiny central peak. If comparing this MHL to that measured for YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\), Bi-2212 and underdoped LSCO \((x = 0.092, T_c = 26 \text{ K}, \text{ shown in fig. 3(b)})\), one can clearly see that the central peak becomes very small or disappears for the overdoped LSCO \((x = 0.24)\) sample.

For our present overdoped LSCO single crystal \((x = 0.24, T_c = 25 \text{ K})\), it is known that \(t_c = T_c/T_{c\text{opt}} = 25 \text{ K}/38 \text{ K} = 0.66\), thus the sample is close to the heavily overdoped region. In fig. 4(a) and (b), we present the MHLs for a Tl-2201 single crystal \((T_c = 35 \text{ K})\) with \(t_c = T_c/T_{c\text{opt}} = 35 \text{ K}/95 \text{ K} = 0.37\), thus it is in the heavily overdoped regime. In the low-temperature region, a penetration peak is observed at a rather low field. This penetration peak at a low field results from the relatively strong coupling between the tiny superconducting islands in the low-temperature region. When the temperature is increased, this first peak disappears quickly (as shown in fig. 4(b)) and a second peak at a higher field develops. This again shows the easy penetration of magnetic flux in the heavily overdoped region.

Next we present further evidence by showing the dynamical flux penetration process. According to our picture, the bulk superconducting state is established by the Josephson coupling between the tiny superconducting islands. In the short time scale the high slope of flux gradient \(\frac{dB(x)}{dx}\) corresponding to a large supercurrent can be maintained since the state relaxes only a little from the initial state \((t = 0, \text{ the moment the field sweeping is just stopped})\). A long time relaxation process will drive the system closer to the static state, so that the central penetration peak will become more and more invisible. In order to check this
Fig. 4 – MHLs measured for a heavily overdoped Tl-2201 single crystal at (a) temperatures of 4, 6, 8, 10, 14, 24, 34 K (from outer to inner) and (b) at 24 K. In the low-temperature region, a penetration peak appears at a rather low field. This penetration peak at a low field probably results from the relatively strong coupling between the tiny superconducting islands in the low-temperature region. When the temperature is increased, this first peak disappears as shown in (b).

Fig. 5 – Time-dependent MHLs for the overdoped LSCO single crystal \( x = 0.24 \), \( T_c = 25 \) K. It is clear that the magnetization at zero field relaxes much faster than that at a high field leading to the vanishing of the central penetration peak. The inset gives a schematic view for the percolating superconductivity in the heavily overdoped regime. The rounded squares and the ellipses stand for the tiny superconducting islands and the Josephson vortices, respectively.
systems with very different structural details. For example, in LSCO single crystals there is a structural phase transition from orthorhombic to tetragonal at a certain temperature, while in Tl-2201, Bi-2201 and Bi-2212 systems no such transition is expected. In addition, the doping to the overdoped region in the LSCO, Bi-2201 and Y(Ca)-123 systems is by cation substitution, while doping in Tl-2201 and Bi-2212 is through oxygen post-annealing which will induce rather different structural deformations. For a Bi-2212 single crystal with $T_c = 90$ K, the MHL has been measured and shown in fig. 2. When this sample is post-annealed in high-pressure O$_2$ (100 atm, 500 °C), the transition temperature $T_c$ drops to 72 K and the central peak becomes much smaller than the optimally doped sample. Similar data were published recently [13], although no definite reasons were given. It would be very interesting to see whether this central peak will vanish completely when further doping Bi-2212 to heavily overdoped regime. Since this effect has been found in five systems: LSCO, Tl-2201, Bi-2201, Bi-2212 and Y(Ca)-123, we anticipate that whenever a HTS is driven to the heavily overdoped region it may be possible to observe this effect.

The second argument would be that this easy-penetration effect is induced by the very low superfluid density (thus low critical current density and irreversibility line), although the electronic property is uniform [14] throughout the whole sample. This argument gets, however, no support from the data measured for any underdoped HTS sample which has also a very low superfluid density, but the penetration peak can be easily observed. As mentioned already, this easy-penetration effect has never been observed in underdoped HTS samples. Therefore, even for a superconductor with very low superfluid density one should be able to observe a penetration peak when the superconducting state is uniform.

This easy-penetration effect can get an explanation from the picture based on the electronic phase separation in a heavily overdoped HTS sample. The resulting phase consists in many tiny superconducting islands surrounded by the hole-rich metallic but non-superconducting regions [4, 7]. The bulk superconducting state is established by the Josephson coupling or proximity effect between these islands. In this sense the critical current density (or vortex pinning force) is much smaller in the heavily overdoped region than in the underdoped region, although the superconducting transitions occur at almost the same temperature [15]. It should be noted that in the slightly overdoped regime with hole number from 0.15 to 0.19, since the non-superconducting regions are small and separated and act as vortex pinning centers, and probably the superfluid density is also higher here, therefore the critical current density in this region is even higher than in the underdoped or optimally doped region [16]. It has been found that the superconducting criticality for the underdoped and the overdoped sample is the same [17], reflecting probably the same condensation processes, e.g., both are BEC in the two doping limits. A schematic picture for the heavily overdoped sample is shown in the inset of fig. 5. In the low-field region, the magnetic flux (probably the Josephson vortices) will easily penetrate into the center of the sample through these weak-coupling channels. Therefore the central peak will become too small to be observable. Since the decoupling transition coincides with the irreversibility line [4, 15], we would not expect any irreversible flux motion within the superconducting islands. This may indicate that the islands are too small to allow the formation of vortex lattice. If this is the case, we can expect that the islands have a size between several times the coherence length and 1000 Å. When the field is increased, one can observe an increase of the magnetization. This may be induced by the gradual formation of a vortex solid state. However, it is important to note that this vortex solid state, if it exists, is formed by the Josephson vortices rather than by the normal vortices. This certainly deserves a further check.

In conclusion, by measuring the MHLs for several heavily overdoped high-temperature superconductors (LSCO, Tl-2201, Bi-2201, Bi-2212 and Y(Ca)-123), for the first time we have...
observed a new type of MHL characterized by the vanishing of the central penetration peak. This effect has never been observed on underdoped or optimally doped samples and can be attributed to the percolative superconductivity due to the inhomogeneous electronic state in the heavily overdoped region.

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