Anomalous magnetization transition accompanying the irreversibility line in high-temperature superconductors

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By measuring the temperature-dependent diamagnetic moments in the field cooling and zero-field-cooling processes on $Bi_2Sr_2CaCu_2O_8$, $Bi_2Sr_{2-x}La_xCuO_6$ and overdoped $La_{2-x}Sr_xCuO_4$ single crystals, it is found that there are two distinct transitions on each $M(T)$ curve at a high field (above about 1 kOe), but only one transition at a lower field. The unexpected second step on the $M(T)$ curves at the irreversibility point cannot be understood within the picture of a second-order vortex-glass melting. For $YBa_2Cu_3O_{7-\delta}$ single crystals, this anomalous step was not observed up to a field of 5 T revealing a vortex glass transition behavior. Possible reasons are given to interpret this anomaly.

I. INTRODUCTION

The dynamics of vortex matter has received tremendous efforts in the past decade.[1] A typical example is about the flux dynamics and the vortex phase diagram in $Bi_2Sr_2CaCu_2O_8$ single crystals (Bi-2212) on which many interesting phenomena, such as the second peak (SP) on the magnetization hysteresis loop, have been observed. By using 2DEG miniature Hall-probe arrays to detect the local magnetic induction, Zeldov *et al.*[2] derived a reasonable picture for the vortex phase diagram[3]. It was suggested that the SP appearing between 20 and about 40 K in the low-field region is induced by the competition between the surface and/or geometrical barrier and the bulk pinning. With increasing the magnetic field the vortex system will change from the low-field quasilattice (with high mobility) to the highly disordered vortex-glass state (with low mobility) at a high field. Therefore, the irreversibility line (IL) in high-field region has been explained as a continuous vortex-glass transition. Above 40 K the quasilattice will melt through a first-order-transition (FOT) and the IL in that region (above the melting temperature) is thus determined by the geometrical barrier.[4] This picture, though based on solid data and thus recognized by many researchers, leaves, however, still some unsolved problems. One question is, for example, why below about 20 K, the bulk pinning suddenly becomes extremely strong so that the second peak is almost invisible. It seems that there is a threshold temperature for the drastic flux motion when going from low temperature to the SP region as discovered recently by Tonomura *et al.*,[5] although there is no reason to believe that there exists a sharp transition of pinning strength at round 20 K since the elementary pinning force[6] is proportional to $(1-t^2)^n$ with $n>0$ and $t=T/T_c$. In this paper, we present data to show that the IL in the high-field region is accompanied by a clear step on the $M(T)$ curves measured both in the zero-field-cooled (ZFC) and (FC) processes. The unexpected second step at a lower temperature on the FC curve is difficult to understand unless one assumes that the ''superconducting background'' undergoes a transition and the diamagnetic moment due to the Meissner edge current within the penetration depth at the perimeter of the sample drops or even vanishes. By combining this phenomenon with the observed well-known two-dimensional (2D) dislocation networks[7] in this system, we propose a picture to consider the superconducting background as a superconducting glass state which contains many small superconducting islands coupled by Josephson effect (or proximity effect). The major experimental observations can be reasonably explained based on our picture.

II. EXPERIMENT

All the $M(T)$ curves were measured by a Quantum design superconducting interference device (SQUID, MPMS, 5.5 T) with different scan length. The magnetic hysteresis loops with the SP effect were measured by an Oxford Instrument vibrating sample magnetometer (VSM, Model 3001, 8 T) with a resolution of 10^{-6} emu. $Bi_2Sr_2CaCu_2O_8$ (Bi-2212), $Bi_2Sr_{2-x}La_xCuO_6$ (Bi-2201), and overdoped ($x=0.24$) $La_{2-x}Sr_xCuO_4$ (LSCO) single crystals with dimensions of approximately 2×2 mm^2 and thickness around 100 μm have been investigated and the major features remain the same for different samples. Bi-2212 and Bi-2201 single crystals were grown by using the self-flux method, while the LSCO was grown by using the floating-zone technique. The T_c (onset) and the transition width, as determined from the ac susceptibility measurement, are 85.4 and 2 K for Bi-2212, 28 and 2 K for Bi-2201, 26 and 1 K for LSCO, respectively. For all the measurement, the magnetic field was applied parallel to the c axis. For the measurement of $M(T)$, two methods were used, namely zero-field-cooling (ZFC) and field-cooling (FC). In the ZFC process the sample was first cooled in zero field from 120 K to the desired temperature, then an external field was applied and the magnetic moment was measured in the warming up process. In the FC process, the sample was cooled down under a field from 120 K to the desired temperature and the magnetic moment was also measured in the warming up process. The difference between a ZFC and a FC process is that in the ZFC process the magnetic flux enters the sample, in contrast to escaping from the sample in the FC process. It is important to note

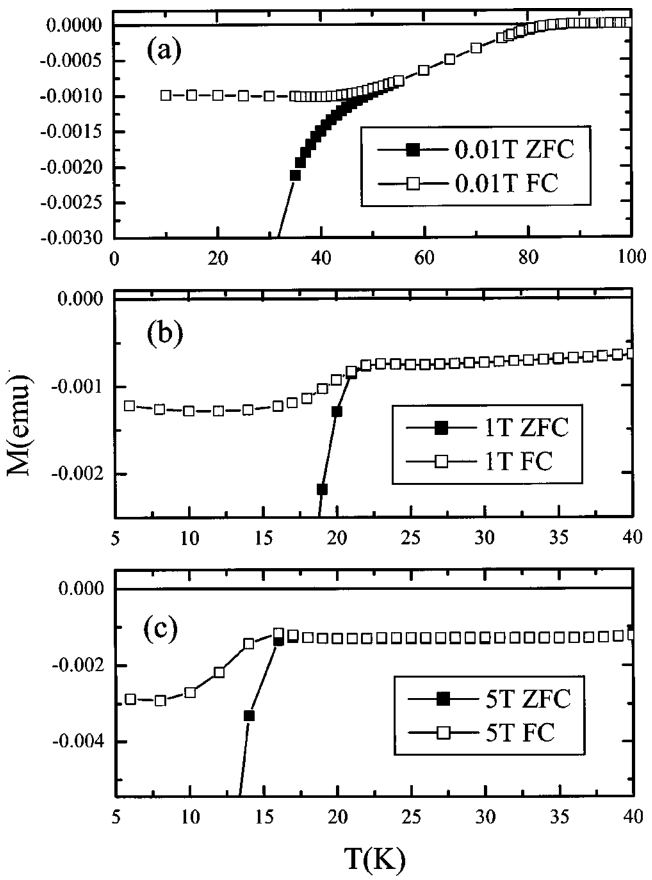

FIG. 1. Temperature-dependent diamagnetic moments measured in the ZFC and FC processes at magnetic fields of (a) 0.01 T, (b) 1.0 T and (c) 5.0 T. An unexpected second step appears clearly on the $M(T)$ curves in the high-field region at the irreversibility point.

that, for the FC method, the data was collected both in the cooling process (FCC) and the warming up (FCW) process. In both FCC and FCW processes, the results are almost the same. For simplicity, in this paper only the data for ZFC and FC(W) are presented.

III. RESULTS

Shown in Fig. 1 are transition curves measured on the Bi-2212 sample under three typical fields $\mu_0 H_{ex}=0.01$, 1, and 5 T, respectively. At 0.01 T, it is easy to find that there is only one transition at $T_c \approx 86$ K, without any trace for a second transition on the FC curve. When the field is increased above about 0.1 T, a second transition on both the ZFC and FC curves emerges and grows monotonically [see Figs. 1(b) and 1(c)]. All the $M(T)$ curves show a rather stable transition near $T_c \approx 86$ K, which can be described very well by the critical fluctuation theory.[8] For simplicity, we show in Figs. 1(b) and 1(c) only the second transition. The diamagnetic moment varies very slowly with temperature above the second transition until it enters the region for the first transition at around 86 K. This anomalous step was observed in more than 10 pieces of samples made by different groups, therefore, we would consider it as a common effect in Bi-2212 system. Actually, as shown below, this effect was also observed in Bi-2201 and LSCO system. It is interesting to note that the irreversibility for flux motion occurs simultaneously at the second step on the $M(T)$ curves. Similar effect was reported as early as in 1991 by Kadowaki *et al.*[9]

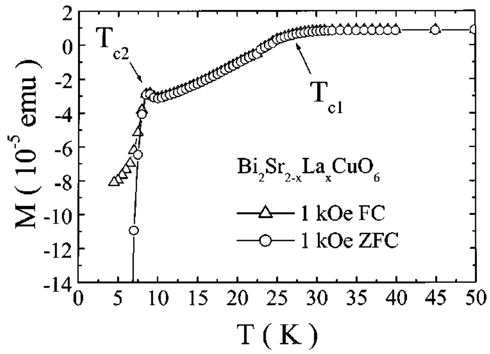

FIG. 2. Temperature dependence of the diamagnetic moments measured in ZFC and FC processes at a field of 1 kOe for one of the $Bi_2Sr_{2-x}La_xCuO_6$ single crystals. Two transitions marked with T_{c1} and T_{c2} can be clearly seen here. Between T_{c1} and T_{c2} the ZFC and FC curves coincide with each other. The irreversibility appears when T_{c2} is reached.

This anomalous second step may be argued as an artifact due to the SQUID measurement, for example, within the scan length, the magnetic field is not perfectly uniform, therefore an anomalous signal may be generated when moving a slab superconductor.[10] This can be, however, ruled out by following arguments: (1) If the second step were induced by the nonuniformity of magnetic field, one should also see it at a low field, which is contradicted to the experimental facts as shown in Fig. 1(a); (2) By changing the scan length from 2 to 8 cm, we observed the same second step on the $M(T)$ curves; (3) A similar step was observed by using a VSM which has only a very short vibrating length; (4) On thick smples instead of thin slabs the second step was still observed.

In order to know whether this anomaly appears also for other systems, we measured $M(T)$ curves on Bi-2201 and LSCO. Surprisingly, a similar effect was observed in Bi-2201 and overdoped LSCO. Shown in Figs. 2 and 3 are the $M(T)$ curves for Bi-2201 and overdoped LSCO, respectively. It is interesting to note that the dimensionality should not play an important role in determining the appearance of

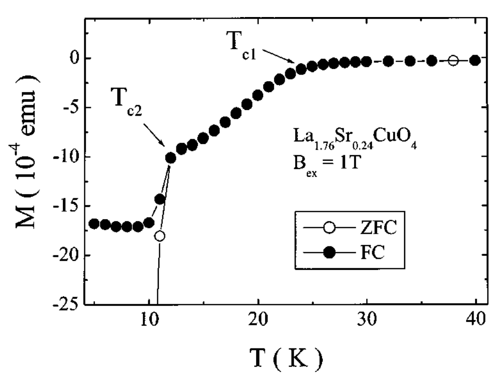

FIG. 3. Temperature dependence of the diamagnetic moments measured for the overdoped $La_{2-x}Sr_xCuO_4$ single crystal sample at an external field of 1 T in the ZFC and FC processes. Two transitions can be clearly seen here. For the underdoped sample there is only one transition which will be presented separately.

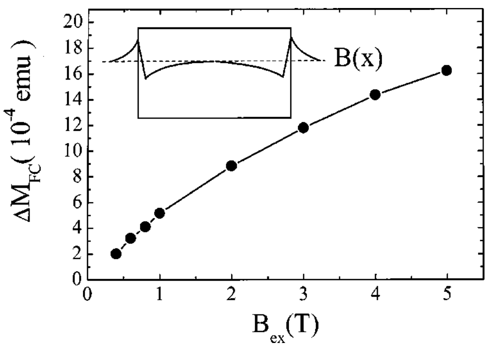

FIG. 4. The jump of the magnetic moment $\triangle M_{\mathrm{FC}}$ on the FC curve before and after the second transition measured for one $Bi_2Sr_2CaCu_2O_8$ single crystal. It is clear that $\triangle M_{FC}$ increases with the external field. The inset shows the field profile during a FC process in which the flux escape from the sample. The magnetic moment in the FC process comprises two terms: the Meissner shielding (negative) and the frozen flux pattern in the interior (positive).

the second step. For Bi-2201 and Bi-2212, the dimensionality $\gamma \approx 50-100$, but for LSCO $\gamma \approx 5$.

The second step on the ZFC curve is relatively easy to understand because enormous flux enters the sample with a drastically fast speed at the irreversibility point and thus the diamagnetic moment drops sharply. Although the steplike behavior, especially that on the FC curve, is related to the irreversibility line (IL) which corresponds probably to the vortex glass (VG) transition, however, the step itself cannot be simply explained as a result of the IL or VG transition. As argued below, probably it is on the other way round, the IL or the VG transition is induced by an effect which is directly related to the step.

IV. DISCUSSION

In order to get a deep insight into the anomaly mentioned above, we turn to the step on the FC curve. This step is very difficult to understand since in the vortex-glass-transition scenario, in the warming up process with the FC mode the frozen sandpile-like flux profile in the interior due to finite j_c will become shallow gradually and eventually flat at the VG transition where $j_c=0$. The diamagnetic moment in the FC process should keep a constant (deep freezing) or slightly increase[11] but should be always continuous, which is in sharp contrast to the experimental observation near the second transition. To better understand this unexpected step on the $M(T)$ curves, we determined the magnitude of the jump $\triangle M_{FC}$ of the magnetic moment at the second transition on the FC curve, that is the difference between the magnetic moments measured at 6 K and just above the transition. As shown in Fig. 4 by the filled symbols, $\triangle M_{\mathrm{FC}}$ increases with the external field. This is qualitatively consistent with what happens in the FC process. The field profile inside the sample in the FC process[12] is shown by the inset in Fig. 4. The edge current due to Meissner effect in the layer of penetration depth provides the diamagnetic moment (DM), while that due to the frozen flux pattern and thus the critical current density j_c in the interior provides the paramagnetic moment (PM). The PM term is always smaller than the DM term, therefore the total magnetic moment is negative. The critical current density which determines the PM will drop with increasing the magnetic field, therefore the higher the magnetic field is, the lower the PM term will be. In short, the increase of the magnitude of the field-cooled magnetization step with applied field (as shown in Fig. 4) arises from the reduction with increasing field of the PM due to flux captured by j_c while the DM due to edge currents is relatively independent of applied field. If there would be no pinning on flux lines, the total magnetic moment should be solely contributed by the edge current due to Meissner effect which should not have any sharp drop since the superconductor does not undergo any kind of transition below T_c. Thus the sharp decrease of the diamagnetic moment measured at the second transition cannot be understood within the picture of a uniform superconductor, which leads to an assumption that the ''superconducting background'' may undergo some sharp transition at the second transition. By combining with the observation of 2D dislocation networks[7] in this system, we propose a picture to consider the superconducting background as a non-uniform state which is constructed by coupled superconductive domains. This non-uniform state is probably induced by the so-called intrinsic inhomogeneous electronic state of high temperature superconductors (HTSC's). In this scenario, at a low field, the superconducting regions are coupled strongly via Josephson effect, therefore one can observe a regular vortex lattice which melts at the melting temperature T_m. At the second transition, the Josephson couplings between the superconducting islands are broken and the edge current is thus interrupted. Above this transition, the diamagnetic signal arises from the individual islands. Since no irreversibility for flux motion above the second transition has been observed, we would assume that each individual island has a size smaller than or equal to the penetration depth which will not allow the formation of a quantized flux line, or each island is very uniform without any defect leading to no pinning effect upon flux lines. In this sense, the real upper critical field H_{c2}, as argued in our former publication,[13] appears at the first transition T_{c1}.

To check this picture, we measured the irreversibility line from the deviating point of the ZFC and FC curves at each field (as shown in Fig. 5 by the filled squares) for one typical Bi-2212 single crystal. As found by many other groups, the IL can indeed be separated into two parts, one part shows a roughly exponential behavior $H_{\mathrm{irr}}(T) \propto \exp(-T/T_0)$ in the high-field region, and another part in the low-field region shows a completely different behavior. What surprises us is that the second step appears only in the high-field region suggesting the same origin as the special form of IL: $H_{\mathrm{irr}}(T) \propto \exp(-T/T_0)$. In the low-field regime, the IL was demonstrated as a strongly geometrical dependent boundary near which is the so-called FOT. In Fig. 5 also shown is the SP line (open circles) which intercepts the IL at a multicritical point. In this paper we focus on the IL in the high-field region. According to our picture, the IL is then corresponding to the field decoupling line, which has been predicted by a recent theory as[14]

$$B_{\mathrm{DC}}=(A/T)\exp(-T/T_0). \tag{1}$$

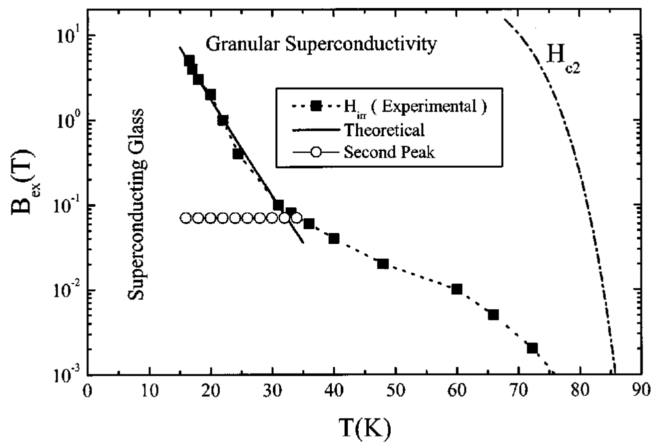

FIG. 5. The vortex phase diagram measured for $Bi_2Sr_2CaCu_2O_8$ samples. The filled squares show the irreversibility line determined from the deviating point on the $M(T)$ curves in the ZFC and FC processes. The unexpected second step appears on the $M(T)$ curve only at a high field accompanied by an irreversibility line $H_{\mathrm{irr}}(T) \propto \exp(-T/T_0)$, showing the same origin for both. The solid line is a fit to Eq. (1) with $T_0=4.5$ K based on the concept of Josephson coupling between the superconducting grains. The second peak line (open symbols) terminates the IL at the multicritical point. The upper critical field line H_{c2} is a schematic show here.

Here A is related to the coupling strength at zero temperature, $T_0=\nu_F/2\pi d$, with ν_F the Fermi velocity of the junction area (nonsuperconducting metal), d is the average distance between the superconducting islands. The solid line in Fig. 5 is a fit to above equation with $T_0=4.5$ K, showing a plausible explanation to the data. Moreover, for $T \gg T_0$, $B_{DC} \propto \exp(-T/T_0)$, which is just the same as what we observed for the irreversibility line. It is interesting to note that, from the data accumulated by Schiling *et al.*,[15] this type of irreversibility line seems to be a common behavior for HTSC's in the high-field region. For example, for $YBa_2Cu_3O_7$, the IL has this type of behavior when the field is higher than about ~ 8 T, although up to date, no attention has been paid to find out if there is a second step on the $M(T)$ curves at a high field in this system. We measured the $M(T)$ curves at fields up to 5 T without seeing the anomalous step at any field. Recently, it was found that the IL obeys a 3D XY scaling in $YBa_2Cu_3O_7$ single crystals,[16] delivering further support for our argument. In that sense, there may be electronic inhomogeneities among all HTS which may be an intrinsic property due to the special electronic state, for example, the stripe phase in the microscopic scale[17] or the phase separated clusters in the mesoscopic scales.[13] It is interesting to note that the anomalous step was observed in Bi-2212 and Bi-2201 system both in the underdoped and overdoped regions, while it is observed only in the overdoped region for LSCO system. This difference may be induced by the easily formed nonuniformly distributed excess oxygens in Bi-2212 and Bi-2201 systems. In LSCO system, the intercalation of excess oxygen is more difficult, therefore the electronic state is quite uniform in the macroscopic scale in the underdoped region. But in the overdoped region, the electronic state of LSCO system may phase separate into macroscopic domains[18] leading to the appearance of the anomalous step on the $M(T)$ curve. A recent study shows the possible connection between the spatial electronic inhomogeneity and the second peak effect.[19]

Regarding the peculiar mixed state properties due to the inhomogeneity in HTSC's, a concept called superconducting glass was proposed shortly after the discovery of HTS's by Müller *et al.*[20] who used this concept to explain the irreversibility line. Theoretically Ebner and Stroud[21] developed a model which consists of many tiny superconducting clusters coupled by weak links and to some extent explained the IL. It is reasonable to consider the vortex system in HTSC's as some kind of glasses [VG or Bose glass (BG)] because of the electronic inhomogeneity. Recently, Gurevich and Vinokur[22] presented a theory concerning the influence of inhomogeneities on the properties in a mixed state. However, as we showed in this paper, the background of this inhomogeneous system may be a superconducting glass, instead of a quasi-uniform superconductor on which the concept VG or BG is based.

For an *s*-wave superconductor, in the Meissner state, there may be no big difference between a superconducting glass or a uniform superconductor because if there is no current flowing in the sample, the superconducting phase can be uniform everywhere. The only difference is that the measured lower critical field reflects only the coupling strength for the superconducting glass instead of a genuine H_{c1} for a uniform superconductor. With increasing the temperature, the uniform superconductor has only one transition that occurs at T_c, while a superconducting glass has two transitions, the first one is also at T_c, but the low temperature one takes place at the decoupling temperature. For a *d*-wave superconductor, there is probably already some spontaneous phase difference across the junction area, thus there is a big difference between these two states. In the mixed state, the intrinsic spatial phase fluctuation may be the origin for the flux pinning. This is probably the reason for Griessen *et al.*[6] to find out that the major pinning in HTSC's is induced by scattering to the superconducting current term of a vortex line. The difference between the mixed states based on a superconducting glass and a uniform superconductor (with pinning defects) can be understood from the different consequences of melting. If a superconducting glass melts, the edge current at the surface drops or vanishes, which is certainly not required for a vortex or a Bose glass.

Finally, we must mention that the phase diagram proposed in Fig. 5 is similar to that suggested by Horovitz *et al.*[23] for $Bi_2Sr_2CaCu_2O_8$ single crystals, the difference is that the Josephson coupling in their picture is between the superconducting Cu-O planes while that in our picture is between the possible superconducting clusters due to intrinsic inhomogeneity of cuprate superconductors. We arrive at this conclusion because the second step on the $M(T)$ curve was observed only on overdoped $La_{2-x}Sr_xCuO_4$ single crystals but not on underdoped samples which normally have even higher anisotropy than the overdoped ones. However, a more elegant experiment for more systems (such as $YBa_2Cu_3O_7$) especially with a high magnetic field, is strongly needed to clarify this issue.

In conclusion, the unexpected steps on the $M(T)$ curves measured in $Bi_2Sr_2CaCu_2O_8$, $Bi_2Sr_{2-x}La_xCuO_6$, and overdoped $La_{2-x}Sr_xCuO_4$ single crystals in the high-field region strongly suggest a picture to consider the superconducting background as a superconducting glass which is probably induced by the intrinsic inhomogeneity of HTSC's. The irreversibility line in the high-field region is thus primarily result from the melting of this superconducting glass. The melting of a VG or a BG, if it occurs, may have only a secondary contribution to the high field IL.

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