The irreversibility line of overdoped Bi$_{2+\delta}$Sr$_{2-(x+y)}$Cu$_{1+y}$O$_{6+\delta}$ at ultra-low temperatures and high magnetic fields

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The irreversible magnetization of the layered high-$T_c$ superconductor Bi$_{2+\delta}$Sr$_{2-(x+y)}$Cu$_{1+y}$O$_{6+\delta}$ (Bi-2201) has been measured by means of a capacitive torquemeter up to $B_a = 28$ T and down to $T = 60$ mK. No magnetization jumps, peak effects or crossovers between different pinning mechanisms appear to be present. The deduced irreversibility field $B_{irr}$ can not be described by the law $B_{irr}(T) \propto (1-T/T_c)^n$ based on flux creep, but an excellent agreement is found with the analytical form of the melting line of the flux lattice as calculated from the Lindemann criterion.

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The investigation of the magnetic properties of high-$T_c$ superconductors (HTS) has attracted a lot of interest, both experimental and theoretical, thanks to the spectacular variety of new and surprising phenomena. The research both on the flux-line statics and dynamics, which has important technological consequences connected to the magnetic irreversibility, and on the fundamental magnetic properties of the superconducting state, like the upper critical field $B_{c2}$ and the subtle nature of the superconducting transition in magnetic field, has been widely developed. These topics are closely related in view of the very intriguing phenomenon of the upward curvature of the upper critical field $B_{c2}(T)$ at low temperatures observed in many HTS [1,2,3,4,5], which is in full contrast with the traditional Werthamer-Helfand-Hohenberg theory [6]. The first suggestion that dissipative measurements tend to yield the irreversibility line rather than the upper critical field dates more than ten years ago [6]. Subsequently, other models have been proposed that focus on the influence of thermal fluctuations [4], vortex lattice melting [5], and low vortex viscosity [10], on the resistive critical field. Other authors pointed out more radical reasons for the upward curvature of $B_{c2}(T)$, like the Bose-Einstein condensation of charged bosons formed above $T_c$ [11], the influence of magnetic impurities [12], or the role of saddle-point singularities in the electron spectrum [12].

The layered cuprate superconductor Bi$_2$Sr$_2$CuO$_6$ (Bi-2201) has been one of the first compounds showing an anomalous temperature dependence of the resistive critical field in a thin film [13]. Subsequent investigations on this compound suffered from difficulties in growing high-quality single crystals. It is only very recently that a study of the DC magnetization has been reported [13]. On the other hand, while sharing most of the structural and physical properties with the other HTS, the low critical temperature of this superconductor allows an experimental investigation of the whole $B-T$ phase diagram at the currently available high magnetic fields and low temperatures. To our knowledge, no extensive studies of the vortex assembly in Bi-2201 have been reported so far. The study of the vortex state in the complete phase diagram is not only interesting in itself, but might have important implications for the anomalous features of the upper critical field.

In this Letter, we investigate the irreversible magnetization of a high-quality overdoped Bi$_{2+\delta}$Sr$_{2-(x+y)}$Cu$_{1+y}$O$_{6+\delta}$ single crystal. The sample was grown from a solution-melt in KCl [14], and has the approximate size of 1100 × 700 × 10 μm$^3$ (mass ~ 0.2 mg). Both the growth method and the small size are advantageous for an extreme homogeneity of the sample. The intrinsic overdoping is due to the Bi excess localized on the Sr positions. The magnetization loops were obtained by means of torque magnetometry, using a very sensitive capacitive torquemeter. We reached temperatures down to $T = 60$ mK in continuous magnetic fields $B_a$ up to 28 T. The field sweep rate $dB_a/dt = 15$ mT/s was chosen in order to have a maximum measuring time for each loop of about one hour with a typical thermal drift smaller than 5 - 10 mK in the whole temperature range. Preliminary measurements were performed in a 10 T superconducting magnet, using $dB_a/dt = 10.8$ mT/s. From our magnetization experiments, we evaluated a critical temperature $T_c \approx 4$ K, in agreement with the overdoping of the sample.

Although we are interested in the irreversibility line for fields applied perpendicular to the $ab$-planes of the crystal, the torque method has no sensitivity for $B_a \parallel c$. The relationship $\tau = M \times B_a$ between torque density $\tau$,
magnetization \( \mathbf{M} \) and applied magnetic field \( \mathbf{B}_a \) suggests that the torque signal can be increased by choosing large values of the angle \( \theta \) between the applied field and the \( c \)-axis of the sample. Actually, in the case of a strongly two-dimensional superconductor like Bi-2201, the scaling analysis in the large anisotropy limit of the Ginzburg-Landau model allows to say that the magnetization \( \mathbf{M} \) lies very close to the \( c \)-axis, while its magnitude is fully determined by the effective field \( B_a \cos \theta \) perpendicular to the \( ab \)-planes. We have chosen \( \theta = 30^\circ \), so that the actual irreversibility field \( B_{irr} \) for \( B_a \parallel c \) is given by \( B_{irr} = B_{irr}^{(a)} \cos 30^\circ \), where \( B_{irr}^{(a)} \) is the applied field corresponding to the vanishing of the magnetic irreversibility. In other words, \( B_{irr} \) is the irreversibility field that we expect to obtain in an ideal experiment with \( B_a \parallel c \) (i.e. with \( \theta = 0^\circ \)), which is not directly measurable with the torque magnetometry technique. From the torque loops, the magnetization can be calculated as \( M = \tau / (B_a \sin 30^\circ) \) but, in view of the arbitrary scaling of the measured torque density, there is no need to take into account the \( \sin 30^\circ \) factor.

The measured torque loops shown in Fig. 1(a) clearly show that the irreversible behavior vanishes quite quickly as the temperature increases. The corresponding magnetization loops shown in Fig. 1(b) have been plotted only for \( B_a > 0.2 \) T, because the division of the torque by the field results in uncertainties for \( B \approx 0 \). Both torque and magnetization loops have a typical shape that scales quite well with temperature. None of our data shows jumps, peaks or fishtail effects. In principle \( B_{irr}^{(a)} \) could be easily determined as the point where the branches for increasing and decreasing field first touch, but we used a more accurate method to obtain \( B_{irr}^{(a)} \) as illustrated in Fig. 2. First, for the difference \( \tau^+ - \tau^- \) of the torque measurements between increasing and decreasing field, respectively, we make a linear fit of what is likely to be the reversible region. Then we define \( B_{irr}^{(a)} \) as the point where \( \tau^+ - \tau^- \) deviates from the fit, helping the eye with a straight line through the irreversible data. For an ideal measurement, the linear fit of \( \tau^+ - \tau^- \) in the reversible region should be a constant equal to zero. However, because the capacitance of the torquemeter is slightly temperature dependent, the thermal drift leads to a non-zero slope for \( \tau^+ - \tau^- \). With the thermal stability of our experiments, that slope is actually extremely small (see the scales of the inset in Fig. 2). But also the irreversible signal vanishes very smoothly for increasing field, and neglecting the nonzero slope would lead to a much higher uncertainty in the evaluation of \( B_{irr}^{(a)} \). Furthermore, this procedure becomes quite useful for the measurements close to \( T_c \), where the signal-to-noise ratio gets worse. Already above \( T \approx 3 \) K the irreversible torque signal, although clearly present, becomes so small that a reliable determination of \( B_{irr}^{(a)} \) is no longer possible. Above \( T \approx 4 \) K we found no more signs of hysteretic magnetic behavior.

Fig. 3 shows the irreversibility line \( B_{irr}(T) \) of our Bi-2201 sample, with a good overlap between the independent sets of measurements at \( dB_a / dt = 15 \) mT/s (resistive magnet) and at \( dB_a / dt = 10.8 \) mT/s (superconducting magnet). We tried to fit our data with the very common function

\[
B_{irr}(T) = B_{irr}(0) (1 - T/T_c)^n
\]

keeping \( T_c = 4 \) K as a fixed parameter. This function has proved to describe many experimental data with \( n \approx 1.3 \pm 0.5 \) and can be qualitatively justified in a flux creep picture, predicting \( n = 3/2 \) or 4 /3 (the value of \( n \) depends on the approximations used to evaluate the pinning energy). In our case, a fit of the data using Eq. (1) with \( n = 1.5 \) is not possible, while a good behavior is only attained for \( n = 5.2 \) (not shown) which has no physical meaning. This is not surprising, because Eq. (1) is obtained by supposing the flux motion to take place for thermal activation over bulk pinning barriers.
Such a model works quite well in disordered systems like Ba$_{1-x}$K$_x$BiO$_3$ [18] but it is less suitable for very clean superconductors. Actually, the vanishing of the irreversible magnetization can also be ascribed to the melting of the vortex solid [19]. For a 3D anisotropic vortex lattice, an useful form of the melting line $B_m(T)$ [10,20] has been obtained making use of the Lindemann criterion, i.e. assuming that the lattice melts when the mean-squared amplitude of the thermal vortex fluctuations $\langle u^2 \rangle_{th}$ exceeds a certain fraction $c_L$ of the vortex spacing. The melting line takes the form

$$B_m(T) = B_{c2}(0) \frac{4\vartheta^2}{1 + \sqrt{1 + 4\vartheta T/T_\circ}}$$  (2)

where $\vartheta = c_L \sqrt{\beta_m/Gi(T_c/T - 1)}$, $T_c = T_c c_L^2 \sqrt{\beta_m/Gi}$, $c_L$ is the Lindemann number, $Gi = 2 \left( \frac{\kappa T_c}{(\pi/\mu_0)B_{c2}(0)^2} \right)^{1/2}$ is the Ginzburg number, $\gamma = \sqrt{m_c/m_{ab}}$ is the anisotropy and $\beta_m \approx 5.6$ is a numerical factor. This expression is supposed to be valid over a wide temperature range below $T_c$, since it is calculated taking into account the suppression of the order parameter close to $B_{c2}$. The fit shown in Fig. 3 is made fixing $T_c = 4$ K and leaving $B_{c2}(0)$ and $c_L^2 \sqrt{\beta_m/Gi}$ as free parameters. From our analysis we obtain $B_{c2}(0) = 16.4$ T (yielding $\xi(0) = 45$ Å) and $c_L^2 \sqrt{\beta_m/Gi} = 0.221$. Estimating $\kappa \approx 40$ and taking $\gamma = 350$ [21,22] we find $Gi = 3.3 \cdot 10^{-2}$, and we finally obtain $c_L = 0.13$.

It is worth noting that Hikami et al. [23] have studied the melting of a 3D flux lattice in strong magnetic fields, obtaining a criterion which is equivalent to the Lindemann’s one with $c_L = 0.14$. This could explain why Eq. (2) gives a good description of the data down to the lowest temperatures, i.e. up to the highest fields. Eq. (2) takes into account only the contribution of thermal fluctuations. We can neglect the contribution due to quantum fluctuations of the vortices [24], because its relative strength is given by $Q \propto Q/\sqrt{Gi}$, where $Q = e^2/\hbar d$ ($d$ is the interlayer spacing). Contrary to $Gi$, $Q$ is unaffected by the anisotropy, so in Bi-2201 we expect the thermal fluctuations to be much more enhanced than the quantum ones. Actually, the contribution of quantum fluctuations of vortices should result in a shift of the melting transition towards lower temperatures and fields, which does not agree with the shape of our measured $B_{irr}(T)$.

With respect to the observed irreversible behavior, one should notice that the torque and magnetization data shown in Fig. 1 tend to rule out the existence of transitions between different phases of the vortex solid, since at least for $T/T_c < 0.7$ no jumps or fishtail effects are present (see e.g. [25]). From another point of view, it follows that no crossovers between different pinning mechanisms are present. For the torque magnetometry technique it can be shown [26] that the pinning force density $F_p(B)$ is proportional to the hysteresis of the torque loop (i.e. what we called $\tau^+ - \tau^-$). In Fig. 4 we have plotted the field dependence of the pinning force density at different temperatures, having rescaled $F_p$ by its maximum value $F_p^{(max)}$ and the applied field by the measured $B_{irr}^{(0)}$. All the curves tend to collapse into an unique shape, which is also an indication that the magnetic field corresponding to the pinning force maximum has approximately the same temperature dependence as $B_{irr}(T)$.

From a qualitative point of view, the irreversibility line we measured can be interestingly compared to the resistive critical field measured by Ososky et al. [1] on a...
Bi-2201 thin film. The upward curvature of the critical field obtained in this transport experiment is very different from the saturating low-temperature behavior of $B_{c2}(T)$ of the Werthamer-Helfand-Hohenberg theory [6]. The temperature dependence of the resistively determined critical field is very similar to the irreversibility field reported here, which confirms that flux lattice melting plays a crucial role in the magnetoresistive transitions.

In conclusion, we have measured the irreversibility line of a Bi-2201 single crystal down to $T = 60$ mK and up to $B_a = 28$ T, obtaining a curve that can be fitted with the form predicted by the Lindemann criterion for the melting of the vortex lattice. The magnetization loops do not show any jump or peak effect, and the pinning force maintains the same shape as function of the field throughout the investigated temperature range. Finally, the behavior of $B_{irr}(T)$ obtained here is very similar to the resistive critical field of a Bi-2201 thin film, suggesting that magnetoresistive experiments are likely to be strongly influenced by flux lattice melting.

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