Irreversibility line of overdoped $Bi_{2+x}Sr_{2-(x+y)}Cu_{1+y}O_{6\pm\delta}$ at ultralow temperatures and high magnetic fields

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The irreversible magnetization of the layered high-T_c superconductor $Bi_{2+x}Sr_{2-(x+y)}Cu_{1+y}O_{6\pm\delta}$ (Bi-2201) has been measured by means of a capacitive torquemeter up to $B_a = 28$ T and down to $T = 60$ mK. The deduced irreversibility field B_{irr} is in excellent agreement with the analytical form of the melting line of a three-dimensional (3D) anisotropic flux lattice as calculated from the Lindemann criterion. For a comparison, the applicability of alternative models based on quantum melting, 2D melting, and flux-creep models is discussed. Rescaling the magnetic-field dependence of the pinning-force with the irreversibility field reveals a similar magnetic-field dependence for different temperatures.

I. INTRODUCTION

The investigation of the magnetic properties of high-T_c superconductors (HTSC's) has attracted much interest, both experimental and theoretical, thanks to the spectacular variety of different 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 a magnetic field, has been widely developed. A large choice of theoretical models is available to describe the vortex phases in HTSC's,[1] allowing one to draw accurate B-T phase diagrams. However, particularly in the compounds with $T_c \sim 100$ K, the experimental results usually do not cover the ultra-low-temperature range, where, for example, the detection of vortex phase transitions would require magnetic fields beyond the currently available values. There are several compounds that, while sharing most of the structural and physical properties with the other HTSC's, have low enough critical temperatures to allow an experimental investigation of the whole B-T phase diagram at the currently available high magnetic fields and low temperatures. Among these compounds, the layered cuprate superconductor $Bi_2Sr_2CuO_6$ (Bi-2201) is certainly one of the most interesting ones, because of its fundamental similarities with the Bi- and Tl-based high-T_c materials. Unfortunately, the difficulties in growing high-quality single crystals of Bi-2201 makes experimental work on this compound less developed compared to other high-T_c compounds.

The aim of our work is to obtain experimental information on the irreversible magnetization of Bi-2201 down to very low temperatures $T \simeq 0.01 T_c$ compared to the critical temperature T_c in order to investigate several models of the physical origin of the irreversibility line. To our knowledge, no extensive studies of the vortex assembly in Bi-2201 have been reported so far. It is only very recently that a study of the dc magnetization has been published.[2] A very intriguing result is the anomalous upward curvature of the resistive upper critical field in a Bi-2201 thin film.[3] Our data will provide an interesting qualitative comparison with that experiment.

II. EXPERIMENTAL

The sample we investigated is a high-quality single crystal of overdoped $Bi_{2+x}Sr_{2-(x+y)}Cu_{1+y}O_{6\pm\delta}$, grown from a solution-melt in KCl,[4] and having the approximate size of $1100 \times 700 \times 10\ \mu m^3$ (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 1 h 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 $\mathbf{B_a} \| c$. The relationship $\tau = \mathbf{M} \times \mathbf{B_a}$ between torque density τ, 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 θ 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[1,5] allows us 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 $\theta = 30°$, so that the actual irreversibility field B_{irr} for $\mathbf{B_a} \| c$ is given by $B_{irr} = B_{irr}^{(a)} \cos 30°$, 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 $\mathbf{B_a} \| c$ (i.e., with $\theta = 0°$), which is not directly measurable with the torque magnetometry technique. From the torque loops, the magnetization can be calculated as $M = \tau/(B_a \sin 30°)$ but, in view of the arbitrary scaling of the measured torque density, there is no need to take into account the $\sin 30°$ 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 nonzero 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.

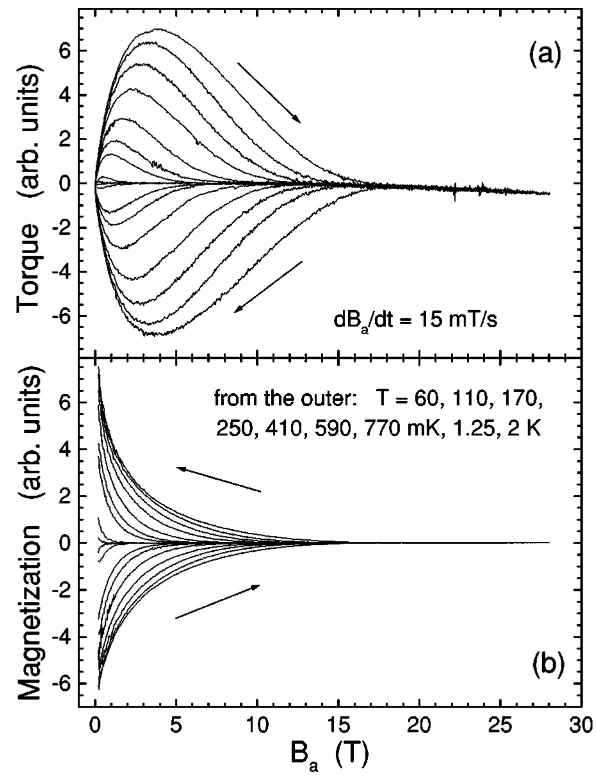

FIG. 1. The torque loops (a) and the corresponding magnetization loops (b) of the Bi-2201 single crystal sweeping the field up and down at $dB_a/dt = 15$ mT/s. Notice the absence of jumps or secondary peaks. The magnetization is plotted only for $B_a > 0.2$ T.

III. DISCUSSION

Figure 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 mag-

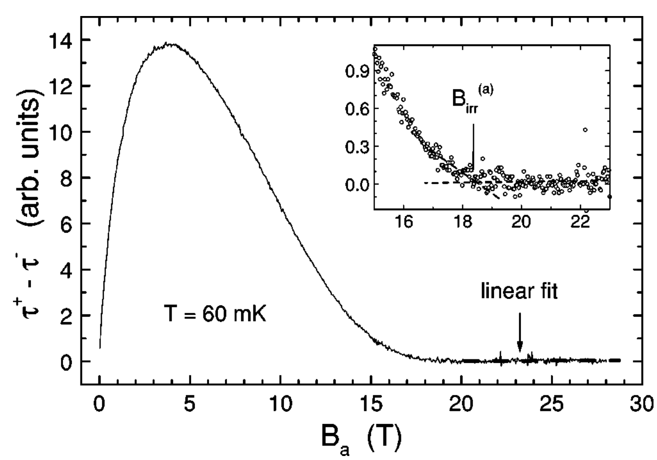

FIG. 2. The difference $\tau^+ - \tau^-$ between the torque recorded for increasing and decreasing field at $T = 60$ mK with a linear fit in the reversible region. The inset shows how $B_{irr}^{(a)}$ can be found, helping the eye with a straight line through the first points in the irreversible region.

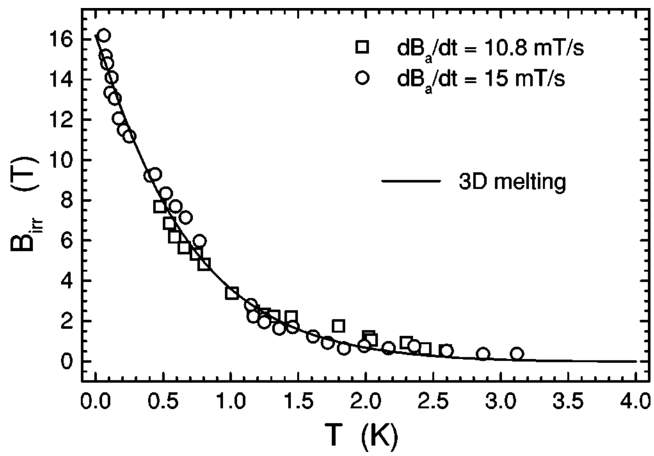

FIG. 3. The irreversibility line $B_{\mathrm{irr}}(T)$ of the Bi-2201 single crystal, recalculated for $\mathbf{B}\|c$ using $B_{\mathrm{irr}}=B_{\mathrm{irr}}^{(a)}\cos 30°$. The data points have been obtained for different sweep rates as indicated. The solid line is a fit through the data for the 3D vortex melting transition using Eq. (1) with deduced $c_{\mathrm{L}}=0.13$.

net) and at $dB_{\mathrm{a}}/dt=10.8$ mT/s (superconducting magnet). The solid line is the best fit to our data, obtained assuming that the vanishing of the magnetic irreversibility corresponds to the melting of the vortex lattice.[6] In particular, we suppose to be dealing with a three-dimensional (3D) vortex lattice (essentially equivalent to a lattice of ''elastic strings''), but where the anisotropic properties of the material are taken into account by the mass ratio $\gamma=\sqrt{m_c/m_{ab}}$. The melting condition can be obtained making use of the Lindemann criterion, i.e., assuming that the lattice melts when the mean-squared amplitude of the vortex fluctuations $\sqrt{\langle u^2\rangle}$ exceeds a certain fraction c_{L} of the vortex spacing a. If we take into account only the effects of thermal fluctuations ($\langle u^2\rangle \equiv\langle u^2\rangle_{\mathrm{th}}$), the melting line takes the form[1]

$$B_{\mathrm{m}}(T)=B_{\mathrm{c2}}(0)\frac{4\vartheta^2}{(1+\sqrt{1+4\vartheta T_{\mathrm{s}}/T})^2}, \qquad (1)$$

where $\vartheta=c_{\mathrm{L}}^2\sqrt{\beta_{\mathrm{m}}/Gi}(T_{\mathrm{c}}/T-1)$, $T_{\mathrm{s}}=T_{\mathrm{c}}c_{\mathrm{L}}^2\sqrt{\beta_{\mathrm{m}}/Gi}$, c_{L} is the Lindemann number, $Gi=\frac{1}{2}[\gamma k_{\mathrm{B}}T_{\mathrm{c}}/(4\pi/\mu_0)B_{\mathrm{c}}^2(0)\xi_{ab}^3(0)]^2$ is the Ginzburg number, $\kappa=\lambda_{ab}(0)/\xi_{ab}(0)$ is the Ginzburg-Landau parameter, and $\beta_{\mathrm{m}}\approx 5.6$ 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 $T_{\mathrm{c}}=4$ K and leaving $B_{\mathrm{c2}}(0)$ and $c_{\mathrm{L}}^2\sqrt{\beta_{\mathrm{m}}/Gi}$ as free parameters. From our analysis we obtain $B_{\mathrm{c2}}(0)=16.4$ T [yielding $\xi_{ab}(0)=45$ Å] and $c_{\mathrm{L}}^2\sqrt{\beta_{\mathrm{m}}/Gi}=0.221$. Estimating $\kappa\sim 40$ and taking $\gamma=350$ (Refs. 7 and 8) we find $Gi=3.3\times 10^{-2}$, and we finally obtain $c_{\mathrm{L}}=0.13$. It is worth noting that Hikami *et al.*[9] 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 $c_{\mathrm{L}}=0.14$. This could explain why Eq. (1) gives a good description of the data down to the lowest temperatures, i.e., up to the highest fields.

At ultra-low temperatures, also the effect of quantum fluctuations of the vortices might be taken into account. The relative strength of quantum fluctuations with respect to the thermal ones can be estimated[10] by a parameter $Q \propto\tilde{Q}/\sqrt{Gi}$, where $\tilde{Q}=e^2\rho_{ab}/\hbar d$ (d is the interlayer spacing); it turns out that quantum fluctuations dominate above the field $B_{\mathrm{Q}}\approx B_{\mathrm{c2}}/Q$. Contrary to Gi, $\tilde{Q}$ is unaffected by the anisotropy, so in Bi-2201 we expect the thermal fluctuations to be much more enhanced than the quantum ones. Nevertheless, evidence for quantum contribution to the flux lattice melting has been reported for the strongly anisotropic κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ organic superconductor,[11] because of the $B_{\mathrm{irr}}\propto T$ law found at low temperatures; our data, instead, show an increasing upward curvature of B_{irr} down to $T=60$ mK.

Another possibility, suggested by the high anisotropy of Bi-2201, is the crossover from three- to two-dimensional vortex system. At sufficiently low temperatures, the flux lines are supposed to decouple in a stack of vortex pancakes,[12] and this obviously influences the features of the melting transition.[13] Above a characteristic crossover field B_{cr}, the melting transition is expected to assume a 2D nature:

$$B_{\mathrm{cr}}\approx 2\pi\frac{\phi_0\ln(\gamma d/\xi_{ab}(0))}{\gamma^2 d^2}, \qquad (2)$$

where ϕ_0 is the flux quantum; in this regime, there is a field-independent melting temperature $T_{\mathrm{m}}^{\mathrm{2D}}$ below which the existence of a 2D vortex solid is expected:

$$T_{\mathrm{m}}^{\mathrm{2D}}\approx\frac{d\phi_0^2}{2\sqrt{3}\mu_0 k_{\mathrm{B}}(4\pi\lambda_{ab}(0))^2}. \qquad (3)$$

With $\gamma=350$ and $\xi_{ab}(0)=45$ Å as before, $d=24$ Å (Ref. 4) and $\lambda_{ab}(0)\sim 1500$–2000 Å, we find $B_{\mathrm{cr}}\sim 0.1$ T and $T_{\mathrm{m}}^{\mathrm{2D}}\sim 30$ K. The deduced combination of these values for B_{cr} and $T_{\mathrm{m}}^{\mathrm{2D}}$ for this model of the 2D cross makes clearly no sense for Bi-2201.

For the detection of the dimensional crossover of the vortex ensemble in $Bi_2Sr_2CaCu_2O_8$, a slightly different approach has been used by Schilling *et al.*[14] Considering a stack of Josephson-coupled layered superconductors and applying the Lindemann criterion $\langle u^2\rangle_{\mathrm{th}}=c_{\mathrm{L}}^2a^2$, the melting line becomes

$$B_{\mathrm{m}}(T)\approx\frac{\phi_0}{d^2\gamma^2}\exp\left(\frac{\phi_0^2c_{\mathrm{L}}^2d}{2\mu_0\lambda_{ab}(0)k_{\mathrm{B}}T}\right). \qquad (4)$$

Our data indeed follow a $B_{\mathrm{m}}\propto\exp(1/T)$ law below $T\approx 0.5$ K, which corresponds to the linear region in $\ln B_{\mathrm{m}}$ vs $1/T$ for $1/T>2$ K^{-1} shown in Fig. 4(a). The solid line is a fit to Eq. (4) with the values for d and $\lambda_{ab}(0)$ as before, and leads to unphysical values $c_{\mathrm{L}}\sim 10^{-3}$ and $\gamma=6$.

So far, we have made the implicit assumption that the irreversibility line (i.e., what we actually measured) coincides with the melting line. This assumption is not straightforward,[15,16] but is particularly acceptable in clean and anisotropic HTSC's. In the opposite case [a typical example is $Ba_{1-x}K_xBiO_3$ (Ref. 17)] the irreversibility line has often been interpreted in a flux-creep picture,[18] supposing the

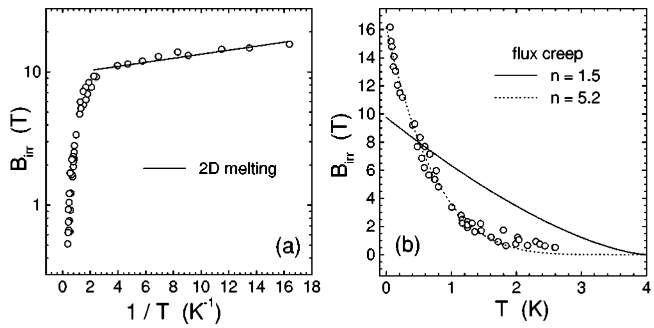

FIG. 4. Other models to interpret $B_{irr}(T)$: (a) the 2D flux lattice melting [Eq. (4)] yields the unrealistic values of $c_L \sim 10^{-3}$ and $\gamma = 6$; (b) the flux creep model [Eq. (5)] requires the unphysical value $n=5.2$ (dashed line), while $n=1.5$ (solid line) does not fit the data.

flux motion to take place for thermal activation over bulk pinning barriers, and obtaining

$$B_{irr}(T)=B_{irr}(0)(1-T/T_c)^n \tag{5}$$

with $n=3/2$ or $4/3$ (the value of n depends on the approximations used to evaluate the pinning energy). Figure 4(b) shows that a fit to Eq. (5) with $n=1.5$ and $T_c=4$ K as fixed parameters is totally wrong, while a good approximation requires the unphysical value $n=5.2$. We take this as a further indication that the irreversibility line in Bi-2201 can be interpreted in terms of flux-lattice melting.

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., Ref. 19). 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[20] 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. 5 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 $B_{irr}^{(a)}$. 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 Osofsky *et al.*[3] on a Bi-2201 thin film. The upward curvature of the critical field obtained in this transport experiment is very different from the saturating

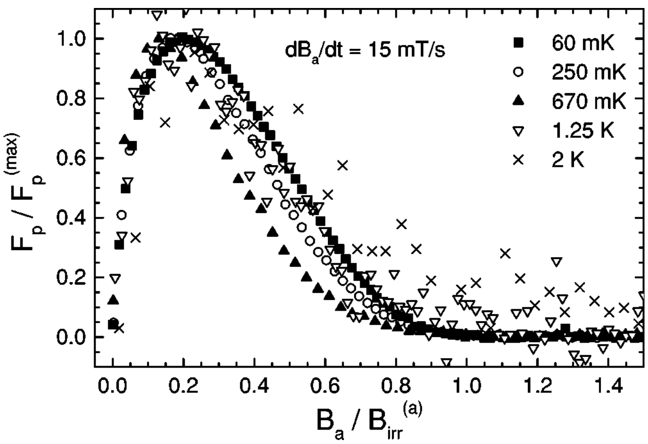

FIG. 5. Pinning force density $F_p \propto \tau^+ - \tau^-$, normalized to the maximum value $F_p^{(max)}$, at various temperatures as a function of the rescaled field $B_a/B_{irr}^{(a)}$. The good overlap and the very similar shapes of all the curves suggest the absence of crossovers between different pinning mechanisms.

low-temperature behavior of $B_{c2}(T)$ of the Werthamer-Helfand-Hohenberg theory.[21] Among the numerous attempts to explain that kind of anomaly,[22–26] it was suggested[27] that magnetoresistive transitions tend to yield B_{irr} rather than B_{c2}. The temperature dependence of the resistively determined critical field in Ref. 3 is actually very similar to the irreversibility field reported here, which confirms that flux-lattice melting plays a crucial role in the magnetoresistive transitions.

IV. CONCLUSIONS

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 a 3D anisotropic vortex lattice. Other models that account for quantum fluctuations, 2D vortex system or flux creep, are unsuitable to describe our data. The magnetization loops do not show any jump or peak effect, and the pinning force maintains the same shape as a 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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