Separated suppression of the transverse and longitudinal Josephson flux mobility in a $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ superconductor

A. R. Jurelo
Departamento de Fisica, Universidade Estadual de Ponta Grossa, Av. Gen. Carlos Cavalcanti 4748, 84030-000, Ponta Grossa, PR, Brazil

P. Pureur and J. Schaf*
Instituto de Fisica, Universidade Federal do Rio Grande do Sul, P.O. Box 15051, 91501-970, Porto Alegre, RS, Brazil
(Received 20 March 2001; published 2 October 2001)

We report on very detailed zero-field-cooling and field-cooling magnetization and magnetoresistance measurements in a polycrystalline $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ superconductor. The results allow us to study the irreversibility line as a function of the magnetic field and temperature. The resistive transition in low fields occurs visibly in two stages, characterizing a granular superconductor. The magnetic irreversibility line as a function of applied field, $T_{irr}(H)$, reveals a flux dynamics with several regimes. In fields below 0.3 kOe, the $T_{irr}(H)$ data define an usual de Almeida–Thouless–like line. However, for fields above this value $T_{irr}(H)$ splits up into two lines of quite different slopes, evidencing a two-step reduction of the flux mobility with decreasing temperatures. We discuss this double onset of irreversibilities in terms of the separated suppression of the transverse and longitudinal fluctuations of the Josephson flux lines. Lacking a specific theoretical approach able to account for the two-step character of the $T_{irr}(H)$ data, we emphasize its striking analogy with the magnetic irreversibility of *Cu*Mn spin glasses and tentatively attribute the upper-temperature line to suppression of the transverse flux mobility along a Gabay-Toulouse-like line and the lower-temperature line to suppression of the longitudinal flux fluctuations along a second de Almeida–Thouless–like line. We suggest that the superconducting glass model including effects from the grain charging energy might be useful to describe these results. We also study the reversible regime of the magnetization above $T_{irr}(H)$ and up to the bulk critical temperature. These results may be interpreted within the three-dimensional XY model for superconducting thermal fluctuations.

DOI: 10.1103/PhysRevB.64.174502 PACS number(s): 74.25.Ha, 74.62.Dh

I. INTRODUCTION

The magnetic behavior of the high-temperature superconductors (HTSC's) as a function of temperature and applied field is largely determined by irreversibilities that depend on the flux mobility and are intimately related to the topology of the superconducting state. Detailed magnetic measurements may thus provide valuable information on the pinning mechanisms as well as on the spatial variation of the condensate wave function.[1] On the other hand, precise magnetoconductivity measurements in the neighborhood of the superconducting transition yield information on the fluctuation regimes and the nature of the superconducting transition. Transport measurements can clearly differentiate between the transition within the grains, which has been called the pairing transition, and the coherence transition which occurs when a phase-coherent state is established in the grain network.[2] Such studies thus are especially suited to reveal details about granular superconductivity.

Superconducting granularity of the HTSC's results not simply from polycrystallinity. Due to the very short coherence length of the Ginsburg-Landau (GL) order parameter, any crystal defect introduces a strong local depression of the superconducting order parameter[3] which may lead to granularity even in single crystals.[4] Extended defects such as dislocations, twinning planes, stacking faults, and others may separate a superconducting grain into subgrains that remain only weakly connected. In the case of $Bi_{2-x}Pb_xSr_2Ca_2Cu_3O_y$, deviations in the oxygen stoichiometry and the introduction of Pb are responsible for the microscopic granularity revealed by studies of fluctuation conductivity in the normal phase.[5] Oxygen depletion also plays an important role. It strongly debilitates the weak links and reinforces the granular character of polycrystalline $YBa_2Cu_3O_{7-\delta}$ (YBaCuO), as verified by resistivity and magnetization measurements.[6] Replacing Ba by Sr in this system strongly increases the twinning[7] and reduces the coherence length,[8] sharpening the topology of the GL order parameter and increasing the granular character of the samples.

Flux dynamics in isotropic and homogeneous low-temperature superconductors (LTSC's) is relatively simple and can be understood in terms of the conventional flux-flow and flux-creep theories.[9] This may even be possible in the case of very pure and homogeneously oxygenated HTSC single crystals.[10] Most of the HTSC samples, however, are granular and the assumptions underlying the conventional theories are not satisfied.

The magnetic behavior of the granular HTSC's in general depends on two quite different contributions to flux dynamics: that of the intergranular, or Josephson, flux and that of the intragranular, or Abrikosov, flux. This is why flux properties in these materials are so complex and also is the most probable reason why the origin of magnetic irreversibilities, although intensely studied since their discovery,[1] is still matter of strong controversy.

Fortunately, in several circumstances the magnetic properties in the granular HTSC's are dominated by only one of the two contributions for the magnetic flux. The fact that the critical field (H_{c1J}) for penetration of Josephson flux into the intergrain spaces is much lower than that for penetration of Abrikosov vortices into the grains themselves (H_{c1g}) means that activation of the Josephson flux occurs for much lower energies than of the intragrain flux. It has been verified that in polycrystalline YBaCuO and YBaCuO/Ag nonrandom composites, effective activation of Josephson flux under low fields occurs even tens of degrees below the magnetic irreversibility limit.[11] Quite generally, the Josephson flux seems to dominate the magnetic behavior of the granular HTSC's in low fields.[8,11–13] On the other hand, for high enough fields and temperatures the motion of the Josephson flux becomes reversible while the Abrikosov flux dynamics still remains irreversible. The Abrikosov flux dynamics has been found to systematically dominate the magnetic irreversibilities of granular HTSC's in a vast high-field region[8,12,13] or even in the whole field range in the case of nearly perfect YBaCuO single crystals,[10,12–19] where the Josephson flux is practically absent.

Another important characteristic of the granular HTSC's resides in the fact that the Josephson flux dynamics is strongly dominated by disorder and frustration in the grain couplings.[20] The de Almeida–Thouless- (AT-) like profile of the $T_{irr}(H)$ data in the low-field region corroborates this fact and was pointed out already in the pioneering work of Müller and co-workers.[1] The AT line[21] is the power law

$$H(T)=H_0^{AT}(1\text{-}t^{AT})^{\alpha} \qquad (\alpha=3/2) \tag{1}$$

where $t^{AT}=T_{irr}^{AT}(H)/T_{irr}^{AT}(0)$. The characteristic field H_0^{AT} and the irreversibility temperature at zero field, $T_{irr}^{AT}(0)$, are fitting parameters. This power law, which was originally determined from mean-field calculations and successfully describes the magnetic irreversibility line of Ising spin glasses,[21] has been found to describe well the low-field $T_{irr}(H)$ data of all the granular HTSC's.[2,6,8,11–13,22–25]

In many irreversibility studies of granular YBaCuO and BiSrCaCuO superconductors[2,6,8,12,13,22–24] it was, however, found that the $T_{irr}(H)$ data, above a crossover field of about 0.5 kOe, systematically deviate from this low-field AT-like line. Moreover, in many cases the behavior of the $T_{irr}(H)$ data above the crossover field[6,8,12,13,22] could be well fitted by a Gabay-Toulouse- (GT-) like power law[26]

$$H(T)=H_0^{GT}(1\text{-}t^{GT})^{\alpha} \quad (\alpha=1/2), \tag{2}$$

where $t^{GT}=T_{irr}^{GT}(H)/T_{irr}^{GT}(0)$ and H_0^{GT} and $T_{irr}^{GT}(H)$ are fitting parameters.

The occurrence of an AT-GT crossover in the magnetic irreversibility line of spin glasses has been long known.[27] Its origin is quite well understood theoretically in terms of an Ising XY or Ising-Heisenberg crossover in the behavior of vector spins.[28,29] This transition occurs when the value of the external applied field reaches a few hundred Oe and becomes much larger than the random local anisotropy fields. The occurrence of such a crossover in the irreversibility line of granular oxide superconductors is, at least, intriguing. In fact, it reinforces the analogy between the magnetic behavior of the granular oxide superconductors and of the spin glasses and raises a number of challenging questions.

In the present work we report on an even much more striking analogy between the magnetic irreversibilities of the HTSC's and spin glasses. We have investigated in great detail the magnetic irreversibility of a polycrystalline $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ [Bi(Pb)-2223] superconductor as a function of the applied field and found, for the first time to our knowledge, suppression of the flux mobility in two stages with decreasing temperature. The *H-T* diagram of the magnetic irreversibilities of this superconductor is almost identical to that of the archetypal *Cu*Mn spin glasses.[27] We also study the field and temperature dependence of the magnetization of our Bi(Pb)-2223 sample in the reversible state above the irreversibility line. These results scale according to the predictions of the three-dimensional (3D) XY model.

II. EXPERIMENTAL METHODS

We prepared a $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ polycrystalline sample using the usual method of solid-state reaction. After thoroughly mixing the precursor powders Bi_2O_3, PbO, CuO, $SrCO_3$ and $CaCO_3$ to the given proportions in an agate mortar, the mixture was reacted at 800 °C during 20 h, and then ground again, pressed into pellets, and annealed at 825 °C for 20 h. After grinding once more and pressing into pellets of 10 mm in diameter and 2 mm in thickness, the samples were sintered at 850 °C during 160 h. All these treatments were performed in air.

dc magnetization measurements were performed as a function of temperature under zero-field-cooling (ZFC) and field-cooling (FC) conditions by using a vibrating sample magnetometer having a sensitivity better than 10^{-4} emu and a temperature resolution of 0.1 K. The magnetoresistance was measured with high resolution using a low-current low-frequency ac experimental setup where a lock-in amplifier is used as a null detector. In this case, temperatures were measured with a Pt resistor corrected for magnetoresistance effects within a resolution of 2×10^{-3} K. Data points are closely spaced so that the temperature derivative of the resistivity, $d\rho/dT$, could be numerically determined.

III. RESULTS

Granular HTSC's having a well-defined superconducting transition temperature within the grains in general shows a two-stage resistive transition. This is particularly visible in $d\rho/dT$ results which displays a sharp peak near T_c and a shoulder in its lower-temperature side.[2] The two-stage resistive transition of our $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ sample is revealed by the results shown in Fig. 1 which were obtained for several applied fields. The prominent peak at 110.12 K is practically coincident with the bulk pairing critical temperature T_C and the shoulder at its left is related to a percolation-like regime where the grains progressively couple until the zero-resistance state is achieved at the coherence transition.[2]

The weak links between the superconducting grains are well known to be very sensitive to magnetic fields.[20] An

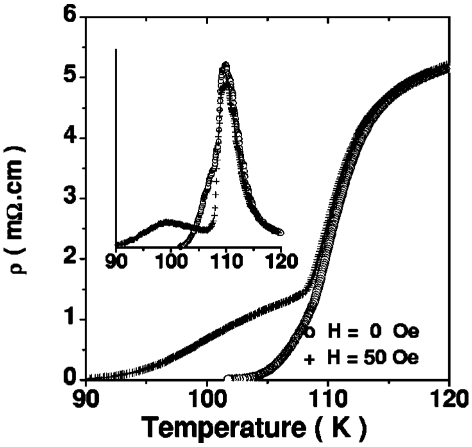

FIG. 1. The resistive transition of our sample for the indicated fields. The inset displays the respective temperature derivatives. Both plots evidence the drastic effects of the small applied field on the grain coupling and coherence transition.

applied field introduces a phase shift in the GL order parameter across the junctions and lowers the grain coupling energy. This is deleterious to the phase coherence and displaces the zero-resistance point to lower temperatures. In Fig. 1 the broadening produced by the applied field appears very clearly in the lowest step of the resistive transition. In the low-field limit, the fluctuation regime precursor to the coherence transition is enlarged.[2,30] The bulk transition, however, remains sharp and practically at the same temperature.

Figure 2 shows some of the dc magnetization measurements in our Bi(Pb)-2223 sample as a function of the temperature in the indicated fields. These experiments were performed under zero-field-cooled (M_{ZFC}) and field-cooled (M_{FC}) conditions for a large number of applied fields up to 5 kOe. The first inflection of the magnetization with lowering temperature denotes T_c, which coincides within the experimental precision with the temperature of the peak in $d\rho/dT$ shown in the inset of Fig. 1. The temperature where the ZFC and FC curves split apart is the irreversibility limit for the given applied field. As shown in Fig. 2, these curves separate tangentially, making it difficult to see where exactly they separate. The irreversibility limit becomes much sharper by plotting the difference $\Delta M = M_{FC} - M_{ZFC}$ as a function of T, as displayed in Fig. 3 for several applied fields. In practice, however, the interpretation of these plots may be complicated by the presence of thermal gradients. For a superconductor, this gradient depends on temperature near T_c, where its thermal conductivity changes considerably. In our experiments we take special care to shortcut temperature gradients and minimize the residual effects by developing correction procedures in the analysis of the data.

The temperature point where the ΔM data abandon the zero base line defines the irreversibility limit of the sample for the respective applied field. Usually, this onset is the only remarkable point shown in the ΔM vs T plots. As depicted in Fig. 3, our ΔM data in low fields effectively display only this characteristic irreversibility point, which shifts from 109 K to about 95 K when the field is increased from 0.003 to 0.3 kOe (see also Fig. 4). The solid line through the low-field

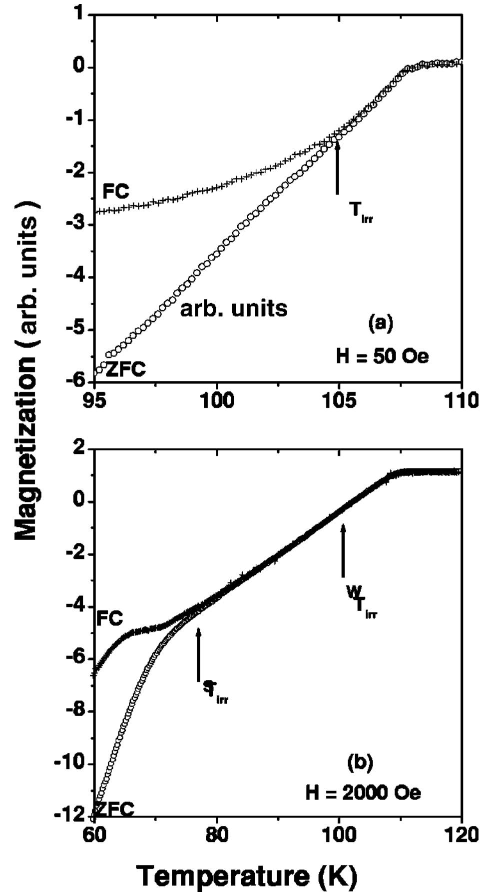

FIG. 2. The ZFC and FC magnetization data as a function of temperature for the indicated fields. The arrows indicate the irreversibility onsets. These limits have in fact been taken from the $\Delta M = M_{FC} - M_{ZFC}$ data in Fig. 3.

irreversibility data, indicated by AT1 in Fig. 4, is a fit to Eq. (1) which leads to $H_0^{AT1} = 8$ kOe and $T_{irr}^{AT1}(0) = 107.5$ K.

From 0.5 kOe and up to the highest fields, $\Delta M(T)$ systematically show an upward inflection point at a temperature neatly below the onset of irreversibility, as shown in the representative results of Fig. 3. The temperatures of the onset (irreversibility limit) and of the inflection point are indicated by arrows and are denoted as T_{irr}^W and T_{irr}^S, respectively, in Fig. 3 as well as in the ZFC and FC magnetization results of Fig. 2. The inflection point T_{irr}^S is obtained from the departure of the ΔM points from the linear fit describing the data between T_{irr}^W and T_{irr}^S, as shown in Fig. 3.

We notice the striking resemblance of Fig. 3 with similar data obtained by Kenning *et al.*[27] for the archetypal *Cu*Mn spin glass. As do these authors, we associate T_{irr}^W with the onset of weak irreversibility effects, which are hardly detectable in plots such as those in Fig. 2. On the other hand, T_{irr}^S is located in a region where the difference between the ZFC and FC magnetization becomes significant. Thus, following again authors in Ref. 27, we attribute T_{irr}^S to the onset of strong irreversibility effects.

Plotting the double breaks T_{irr}^W and T_{irr}^S onto the *H*-*T* diagram of Fig. 4, a consistent feature arises that closely resembles the two-stage suppression of the transverse and

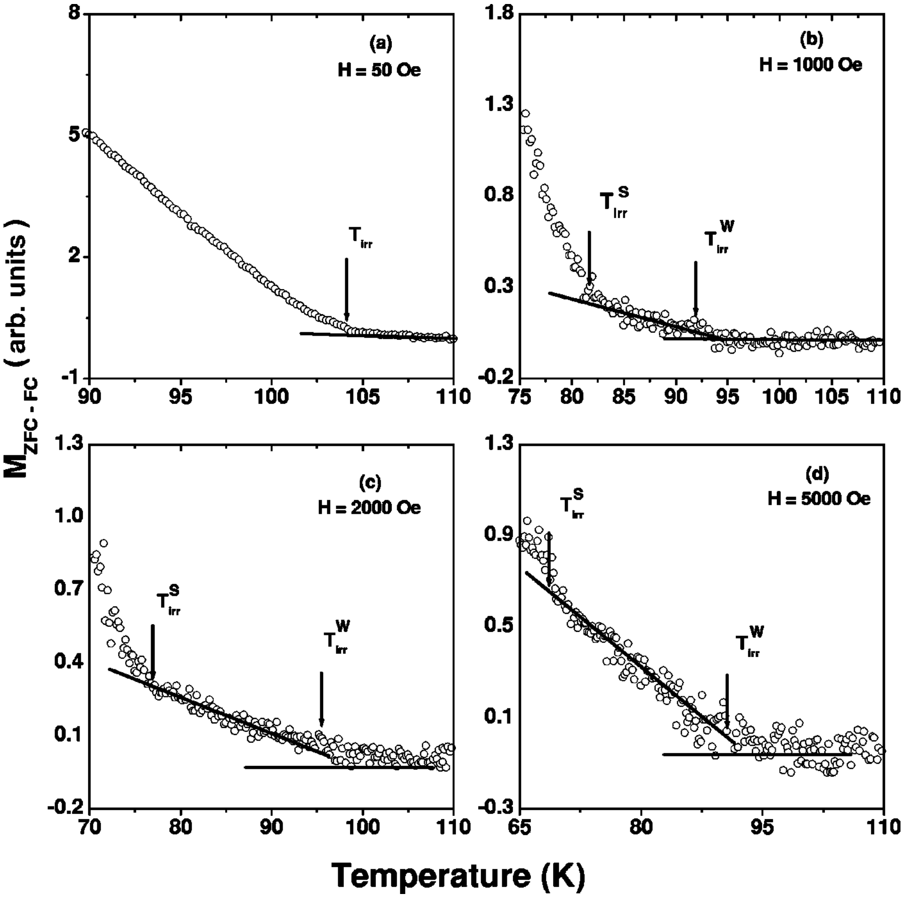

FIG. 3. The $\Delta M = M_{FC} - M_{ZFC}$ data curves for the indicated fields. The arrows indicate the weak and strong irreversibility limits. Note the sharpness of these limits compared to those in Fig. 2.

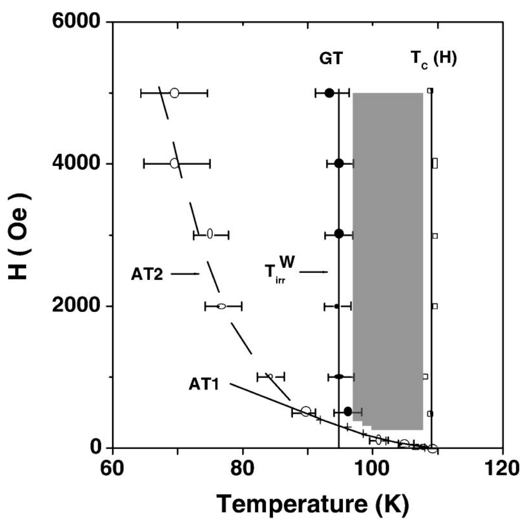

FIG. 4. Open and solid circles are the irreversibility data from ZFC and FC magnetization as a function of applied field. Crosses in the low-field region are the irreversibility data from magnetoresistance. Open squares indicate the $T_c(H)$ values. The solid line through the low-field data and the dashed line, indicated, respectively, by AT1 and AT2 are fittings with a de Almeida–Thouless–like power law [Eq. (1)]. The solid line indicated by GT is a fitting with a Gabay-Toulouse-like power law. The shaded area between the irreversibility and the H_{c2} lines represents the region where the magnetization of the sample is reversible and where the diamagnetic response is genuinely critical (see Fig. 5).

longitudinal spin degrees of freedom which is theoretically predicted[26,29] and experimentally found[27] to occur in spin glasses. In our view, the characteristic temperatures T_{irr}^{W} and T_{irr}^{S} denote a two-step suppression of the flux mobility in our $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_y$ sample. The dashed line through the lower-temperature data branch in Fig. 4, indicated by AT2, is also a de Almeida–Thouless–like line [Eq. (1)], with parameters $H_0^{AT2} = 27$ kOe and $T_{irr}^{AT2}(0) = 95$ K. Note that this line, in spite of the large uncertainties in its determination, in no way can be attributed to the minority phase Bi(2212), which is practically absent in our sample and hardly could have been detected by our experimental method. The solid line through the upper-temperature branch in Fig. 4, indicated by GT, is a Gabay-Toulouse-like line [Eq. (2)] with fitting parameters $H_0^{GT} = 42.75$ kOe and $T_{irr}^{GT} = 95$ K. Although the fittings AT2 and GT are poor, we believe that they are qualitatively correct.

Our present results agree qualitatively well with those of Ref. 22 in the whole range of applied fields. In these previous works, however, the irreversibility line is displaced to lower temperatures and the low-temperature AT2 branch was not found. They also are in good qualitative agreement with the irreversibility data for oxygen-depleted YBaCuO (Ref. 6) and for Sr-doped YBaCuO single crystals and polycrystalline samples.[8,12,13] Some authors[24,25] argue that melting lines, instead of the power-law behavior given by Eq. (1), are found in samples of the same compound as ours but differing in their grain arrangements. The authors of Ref. 24 studied the reversible magnetization on a *c*-axis-oriented bulk sample, while those of Ref. 25 used ac susceptibility and dc magnetization methods on a powder-in-tube sample. The fact that the melting line fitted in Ref. 24 extrapolates to a temperature well above T_c when $H\rightarrow 0$ is a puzzle. Interestingly enough, by imposing that the fitted irreversibility line should vanish at temperatures not higher than T_c, we could fit well the data of Ref. 24 with the power law given by Eq. (1).

We also analyzed the critical behavior of the reversible diamagnetic magnetization M_{rev} of our sample as a function of temperature in the interval between T_{irr}^{W} and T_c. As the paramagnetic contribution above T_c, $M_{T>T_c}$, is very small and constant near the superconducting transition, we have determined M_{rev} as

$$M_{rev}=M_{T<T_c}-M_{T>T_c}, \qquad (3)$$

where $M_{T<T_c}$ is the total measured magnetization below T_c.

We analyze the data by presuming that M_{rev} vanishes at T_c according to a power law of the form

$$M_{rev}=M_0(H)|\epsilon|^{\beta}, \qquad (4)$$

where $\epsilon=(T-T_c)/T_c$, β is a critical exponent, and $M_0(H)$ is a field-dependent amplitude. Then, as in the Kouvel-Fischer method for analyzing critical phenomena,[31] we determine numerically the quantity

$$Y=-\frac{d}{dT}\ln(M_{rev}). \qquad (5)$$

According to Eq. (4), we obtain

$$Y^{-1}=\frac{1}{\beta}(T_c-T). \qquad (6)$$

Then, if a linear behavior is identified along a significant temperature interval in plots of Y^{-1} vs T, the exponent β and the critical temperature T_c can be simultaneously determined. Once β and T_c are known, the amplitude $M_0(H)$ may be determined from Eq. (4). In Fig. 5 we show examples of Y^{-1} vs T plots for several applied fields. All such plots may be fitted to Eq. (6) along a vast temperature interval, and the fitted exponent is $\beta=1.00\pm0.05$. Extrapolation of the fit to $Y^{-1}=0$ leads to a field-independent T_c, which value is in good agreement with the one obtained from resistivity measurements. We notice that the behavior of M_{rev} according to the power law given by Eq. (4) is strictly restricted to the region in the *H*-*T* plane delimited by the irreversibility $T_{irr}(H)$ and H_{c2} lines (see dashed area in Fig. 4). This is precisely the region where the magnetization of our sample is reversible and should be described as an equilibrium property.

Although the value obtained for β is the same as predicted by the mean-field theory,[32] a consistent description of our M_{rev} data requires an interpretation within the 3D *XY* critical thermodynamics. According to Salomon *et al.*,[33] the reversible magnetization near T_c should be written as

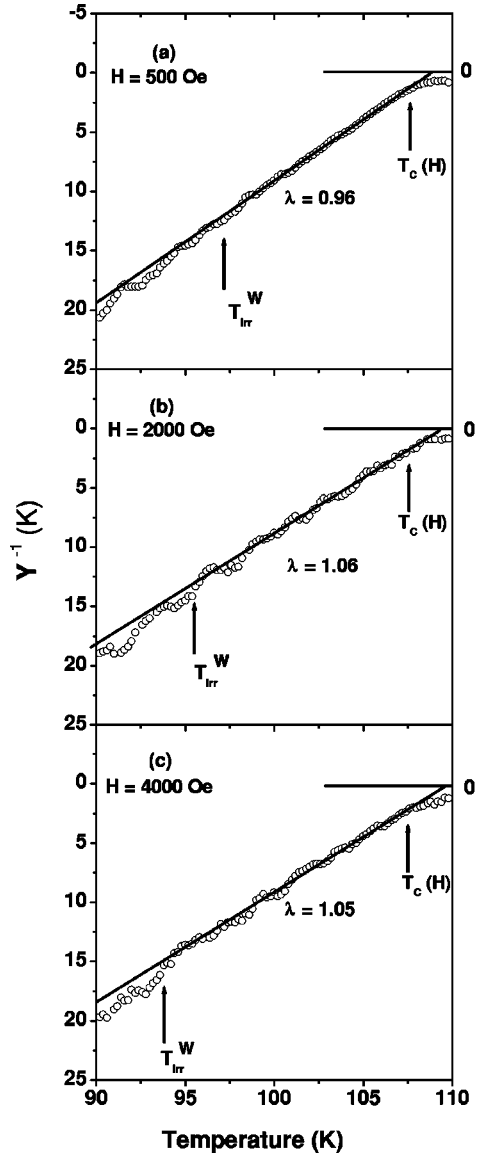

FIG. 5. Plots of the inverse of the logarithmic temperature derivative of the magnetization vs temperature data for the indicated applied fields. The solid lines are fittings to Eq. (6). The arrows indicate the weak and strong irreversibility limits.

$$M_{rev}(H,T)=H^{1/2}F(|\epsilon|/H^{1/2\nu}), \qquad (7)$$

where F is a scaling function and ν is the critical exponent for the coherence length. In the asymptotic limit, Eq. (7) may be simplified as

$$M_{rev}=A|\epsilon|/H^{y}, \qquad (8)$$

where A is a constant and $y=(\nu^{-1}-1)/2$. Equation (8) implies that the amplitude M_0 in Eq. (4) should depend on the field as H^{-y}. Our $M_0(H)$ data could be very well fitted to Eq. (8) and we obtain $y=0.25\pm0.02$. From this value we deduce that $\nu=0.67\pm0.02$, which is quite precisely the value

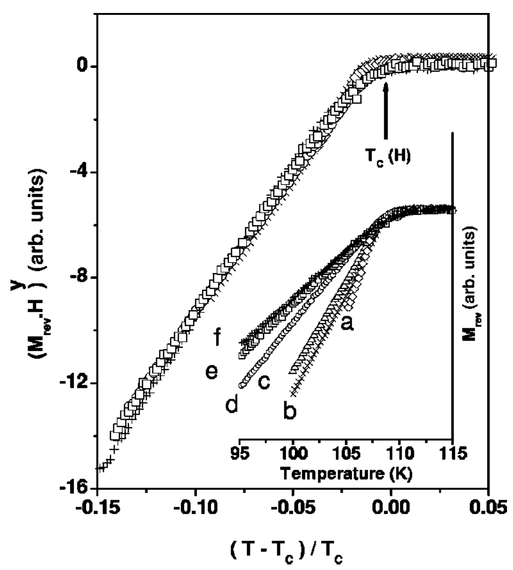

FIG. 6. Scaling of the (FC) reversible magnetization M_{rev} data for (a) 50 Oe, (b) 200 Oe, (c) 500 Oe, (d) 1000 Oe, (e) 3000 Oe, and (f) 6000 Oe (see inset) according to Eq. (8). The data all collapse on the same curve, leading to $y=0.25\pm0.02$ and $\nu=0.67\pm0.02$.

predicted by the 3D XY universality class.[34] In Fig. 6 the M_{rev} results are scaled according to Eq. (8). A good collapse of the data onto a single curve is indeed obtained.

IV. DISCUSSION

The novel feature in the *H*-*T* diagram in Fig. 4 is the double irreversibility onset above the crossover field of 0.5 kOe. This characteristic makes even more neat the striking resemblance between the $T_{irr}(H)$ irreversibility line of granular HTSC's in the low-field limit and the analogous results in the archetypal CuMn spin glass.[27] In particular, the AT- and GT-like behaviors of $T_{irr}(H)$, indicating the onset of weak and strong irreversibility effects, respectively, may be interpreted as signatures of the relevance of disorder and frustration, as in the glassy magnetic systems. The results in Fig. 4 strongly indicate that the superconducting-glass model[20] is a possible source of theoretical understanding for the magnetic behavior of the granular HTSC's at low applied fields. A superconducting glass may be conceived as a highly inhomogeneous superconductor or simply as a disordered and weakly coupled aggregate of superconducting grains under applied field. This model emphasizes the jagged topology of the relevant phase space, which is due to disorder and frustration, and predicts a transition to a glassy and magnetically irreversible superconducting phase when grains become coupled. However, in its simple version,[20,35] the superconducting-glass model predicts that the critical line is completely smooth and thus unable to account for the details of our irreversibility lines.

A possible interpretation within the superconducting-glass model for the AT-GT crossover often observed in the irreversibility line of granular HTSC's at low fields was proposed by Rodrigues, Jr. *et al.*[6] According to these authors, this crossover is related to a charge energy term which should be added to the usual Hamiltonian for a disordered array of Josephson junctions.[20,35] The relevance of a charge energy term for describing the phenomenology of the granular HTSC's was emphasized by several authors.[36–38] Thus, an irregular and weakly coupled aggregate of superconducting grains should be described in terms of the effective Hamiltonian

$$H=-2e^2\sum n_i n_j C_{ij}^{-1}-\sum_{i,j} J_{ij}\cos(\theta_i-\theta_j-A_{ij}). \quad (9)$$

Here the first term on the right-hand side represents the Coulomb energy of the charges in the grains, where C_{ij} are the elements of the capacitance matrix and n_i (n_j) is the number of pairs on grain i (j). The second term is the Josephson coupling term, where the J_{ij} are the coupling energies between neighboring grains *i* and *j* and $\theta_i-\theta_j$ is the difference of the respective order parameter phases. The n_i and θ_j are canonically conjugate variables satisfying the commutation rule $[n_i, e^{\theta_j}]=\delta_{ij}e^{\theta_j}$. The gauge factors A_{ij} are given by

$$A_{ij}=\frac{2\pi}{\phi_0}\int_i^j \vec{A}.\vec{dl}, \quad (10)$$

where ϕ_0 is the elementary flux quantum, $\vec{A}$ is the vector potential, and the line integral is evaluated between centers of grains *i* and *j*. The total phase displacement ΣA_{ij} along closed loops of grains is constrained to $2\pi f$, where f is an integer representing the total number of fluxons enclosed by the loop.

According to the gauge-glass version of the model represented by Eq. (10),[39] frustration is introduced by the randomness in the factors A_{ij}. If π junctions occur between grains concurrently to normal Josephson junctions, frustration may also be introduced by randomness in the couplings J_{ij}. This is the chiral-glass version of model in Eq. (9).[40] The largely varying quality of the junctions, their directional randomness, and the fact that the grains are multiconnected lead to conflict and frustration in the granular arrangement, making it impossible to minimize the system's total energy by simultaneously minimizing the coupling energies of all pairs of grains. Thus, the system loses its ergodicity by freezing into a highly degenerate ground state with a very large number of energy minima of nearly the same value. When the applied field is increased, the randomness of the gauge factors A_{ij} is enhanced and the ability of the aggregate to block the intergranular flux is deteriorated. It is thus easy to understand why $T_{irr}(H)$ diminishes with increasing field.

However, up to now the model of Eq. (9) has not been demonstrated to reproduce the different regimes evidenced by our results. The authors in Ref. 6 suggest that the AT-GT crossover observed in the irreversibility line of slightly underdoped YBaCuO might be due to a single-grain Coulomb energy contribution. This term, reduced to its diagonal components, is formally analogous to a local uniaxial anisotropy field which induces a crossover from Heisenberg to Ising behavior in spin glasses when the applied field is decreased towards zero.[27–29] In the case of granular superconductors, the crossover would be from the XY to an Ising type of symmetry of the order parameter. According to the mean-field theory,[29] in both cases the lowering in the effective dimensionality of the order parameter with decreasing fields would induce a crossover in the freezing temperature from a GT-like line to an AT type of behavior. Moreover, according to this theory, the GT line corresponds to the freezing of the transverse degrees of freedom of the order parameter, whereas the longitudinal freezing would occur at an AT-like crossover line at lower temperatures, giving origin to the double irreversibility line observed in spin glasses.[27] Pushing further the analogy between these magnetic systems and the granular HTSC's, we might suggest that the observed weak (GT line) and strong (AT-2 line) irreversibility limits in our sample are also due to separate freezing of the transverse and longitudinal components of the order parameter, respectively. In the language of vortices, one would say that at the GT line topological vortices, formed by loops of weak-coupled grains enclosing a cross-sectional area with respect to the field orientation, start to be stabilized. At the AT-2 line these vortices acquire longitudinal coherence. In other words, their length becomes comparable to the sample's width. According to this interpretation, it is natural that the AT-2 line does not fit as a mere continuation of the low-field AT-1 line. The former denotes a crossover in a region in the H-T diagram where the order parameter has XY symmetry, while the latter denotes a frontier in a region where the system behaves effectively as Ising like.

The interpretation based on Eq. (9) for the low-field magnetic irreversibilities observed in our granular HTSC's is also favored by the fact that the magnetization scales as predicted by the 3D XY universality class in the reversible regime above the GT and AT-1 lines in the H-T diagram of Fig. 4. This means that vortices exist only as fluctuations in this region, so that the irreversibility line cannot be attributed to depinning effects. On the other hand, the relevance of the Coulomb energy term implies that the size of superconducting grains ranges about 0.1μ.[6] Although being much smaller than the typical crystallite size, this value is qualitatively consistent with the picture that emerges from fluctuation conductivity studies in the same sample.[5] These results revealed that our Bi(Pb)-2223 superconductor behaves as microscopically granular. This state is generated by strong disorder at the atomic level and is characterized by a correlation length larger than the intrinsic coherence length, but much smaller than the metallurgical grain size.[5]

We note, however, that the above-outlined interpretation, based on the spin-glass phenomenology, for describing irreversibilities in the granular HTSC's is subject to criticisms. One of them concerns the strict analogy between the XY spin glass and the superconducting-glass problem represented by Eq. (9). It is clear that the two problems are not identical from a theoretical point of view. From the experimental point of view, some aspects should also be taken into account. It has been shown that the irreversibility line at low fields in granular HTSC's is not a phase boundary. Indeed, a true percolationlike phase transition (coherence transition) involving the whole array of grains occurs at temperatures neatly below $T_{irr}(H)$, according to several magnetoconductivity and specific heat studies.[2,5,41] These results indicate that the irreversibility temperatures correspond to the formation of relatively small clusters with closed loops of Josephson-coupled grains. However, the postulated analogy with the spin-glass physics could remain valid. In spite of being a short-range order effect, the irreversibility phenomenon in granular HTSC's may still be described by Eq. (9), since even relatively small closed loops of grains behave as frustrated structures in the presence of a field.[20,35] According to this view, the irreversibility temperatures are not the equivalent of the critical temperature of the spin-glass mean-field theory, but rather the granular superconductor analog of the Griffiths temperature, where short-range ordering effects become relevant.

The concept of separated transitions for the transverse and longitudinal degrees of freedom has also been incorporated to the description of the Abrikosov flux-lattice melting in layered superconductors.[42] In this case, the vortex solid first melts into a liquid where correlation perpendicular to the field orientation is lost. Then, at a higher temperature, a decoupling related to the planar anisotropy occurs and the vortex lines lose longitudinal coherence. This phenomenon has been evidenced in some high-quality single crystals.[43] We, however, believe that it can hardly explain the magnetic irreversibilities observed in our granular sample, where disorder at a mesoscopic and microscopic level plays an important role. In our low-field experiments, the number of vortices per grain is so small that the concept of a vortex lattice cannot be applied to a single grain. Moreover, the fact that the 3D XY scaling holds for the magnetization in a large reversible regime above the irreversibility line seems to rule out any major effect from the Abrikosov vortices in the region of the H-T plane shown in Fig. 4.

In summary, our experiments make clear that disorder, frustration, and magnetic irreversibilities are intimately related in our granular Bi(Pb)-2223 superconductor. The obtained irreversibility lines are collective grain properties and cannot be attributed to depinning or single-grain melting of Abrikosov flux lines. The superconducting-glass model including the effect of the Coulomb charging energy is suggested as a possible theoretical framework to account for the observations since the irreversibility effects and the obtained $T_{irr}(H)$ diagrams show striking similarities with those of the spin glasses. In this work, in addition to hitting on the unsolved problem of the crossover from the low-field AT-like to the higher-field GT-like regime in the irreversibility line, a completely new problem is raised: the two-step irreversibility onset above the crossover field. We emphasize the similarity of both of these properties with the analogous features of the archetypal CuMn spin glass. This fact leads us to suggest that the AT-GT-like crossover in the $T_{irr}(H)$ lines of our granular HTSC sample occurs when the local phase anisotropies, induced by the Coulomb term of the effective Hamiltonian, Eq. (9), collapse under the external applied field. This is in analogy with a similar phenomenon in spin glasses.[28] The analogy, however, goes much farther. The upper- and lower-temperature branches of our $T_{irr}(H)$ data in Fig. 4 may represent separated suppression of the transverse and longitudinal fluctuations of the magnetic fluxons, respectively. When the first superconducting-grain loops close, they trap Josephson-vortex segments within them, starting suppression of the transverse fluctuations. At a somewhat lower temperature, depending on the applied field, the suppression of longitudinal fluctuations begins as well, when the first vortices percolate through the whole sample.

ACKNOWLEDGMENTS

The authors gratefully acknowledge support from the Brazilian Ministry of Science and Technology under Contract PRONEX/FINEP/CNPQ No. 41.96.0907.00.

*Corresponding author. Mailing address: Instituto de Física–UFRGS, Cx. P. 15051, Av. Bento Gonçalves, 9500–Campus do Vale, 91501-970 Porto Alegre–RS, Brazil. FAX: (5551) 319-1762. Electronic address: Schaf@if.ufrgs.br

[1] K.A. Muller, M. Takashige, and J.G. Bednorz, Phys. Rev. Lett. **58**, 1143 (1987).
[2] J.R. Rojas, R.M. Costa, P. Pureur, and P. Prieto, Phys. Rev. B **61**, 12 457 (2000).
[3] G. Deutscher and K.A. Müller, Phys. Rev. Lett. **59**, 1745 (1987).
[4] M. Daeumling, J.M. Seuntjens, and D.C. Larbalestier, Nature (London) **346**, 332 (1990).
[5] A.R. Jurelo, J.V. Kunzler, J. Schaf, P. Pureur, and J. Rosenblatt, Phys. Rev. B **56**, 14 815 (1997).
[6] P. Rodrigues, Jr., J. Schaf, and P. Pureur, Phys. Rev. B **49**, 15 292 (1994).
[7] K. Saito *et al.*, Phys. Rev. B **58**, 6645 (1998).
[8] V.N. Vieira, P. Pureur, and J. Schaf, Physica C **353**, 241 (2001).
[9] P.W. Anderson and Y.B. Kim, Rev. Mod. Phys. **36**, 39 (1964).
[10] Y. Yeshurun and A.P. Malozemoff, Phys. Rev. Lett. **60**, 2202 (1988).
[11] J.P. da Silva, J. Schaf, P. Pureur, and S. Reich, Physica C **332**, 31 (1999).
[12] V.N. Vieira, J.P. da Silva, and J. Schaf, Physica C **341**, 1155 (2000).
[13] V.N. Vieira, P. Pureur, and J. Schaf, Physica C **354**, 299 (2001).
[14] G. Blatter and B. Ivlev, Phys. Rev. Lett. **70**, 2621 (1993).
[15] A. Shilling, H.R. Ott, and Th. Wolf, Phys. Rev. B **46**, 14 253 (1992).
[16] H. Safar, P.L. Gammel, D.A. Huse, D.J. Bishop, J.P. Rice, and D.M. Ginsberg, Phys. Rev. Lett. **69**, 824 (1992).
[17] G.T. Seidler, T.F. Rosenbaum, D.L. Heinz, J.W. Downey, A.P. Paulikas, and B.W. Veal, Physica C **183**, 333 (1991).
[18] N.E. Hussey, H. Takagi, N. Mori, N. Takeshita, Y. Iye, S. Adachi, and K. Tanabe, J. Supercond. **12**, 583 (1999).
[19] D.A. Cardwell, N. Hari, W. Lo, and A.M. Campbell, Supercond. Sci. Technol. **13**, 646 (2000).
[20] W.Y. Shih, C. Ebner, and D. Stroud, Phys. Rev. B **30**, 114 (1984); **31**, 165 (1985).
[21] J.R.L. de Almeida and D.J. Thouless, J. Phys. A **11**, 983 (1978).
[22] J. Schaf, P. Pureur, and J.V. Kunzler, Prog. High Temp. Supercond. **25**, 262 (1990).
[23] J. Schaf, P. Pureur, and J.V. Kunzler, Phys. Rev. B **40**, 6948 (1989).
[24] Y. Kopelevich, S. Moehlecke, and J.H.S. Torres, Phys. Rev. B **49**, 1495 (1994).
[25] L. Ghivelder, I.A. Castillo, S. Salem-Sagui, Jr., R. de Andrade, Jr., M. Dhallé, G. Grasso, F. Marti, and R. Flükiger, Supercond. Sci. Technol. **12**, 566 (1999).
[26] M. Gabay, and G. Toulouse, Phys. Rev. Lett. **47**, 201 (1981).
[27] G.G. Kenning, D. Chu, and R. Orbach, Phys. Rev. Lett. **66**, 2923 (1991).
[28] G. Kotliar and H. Sompolinsky, Phys. Rev. Lett. **53**, 1751 (1984).
[29] K.H. Fisher, Z. Phys. B: Condens. Matter **60**, 151 (1985).
[30] F.W. Fabris, J. Roa-Rojas, and P. Pureur, Physica C **354**, 304 (2001).
[31] J.S. Kouvel and M.E. Fisher, Phys. Rev. **136**, A1616 (1964).
[32] G. Blatter, M.V. Feigel'man, V.B. Geshkenbein, A.L. Larkin, and V.M. Vinokur, Rev. Mod. Phys. **66**, 1125 (1994).
[33] M.B. Salomon, J. Shi, N. Overend, and M.A. Howson, Phys. Rev. B **47**, 5520 (1993).
[34] J.C. Le Guillou and J. Zinn-Justin, Phys. Rev. B **21**, 3976 (1980).
[35] D. Stroud, and C. Ebner, Physica C **153-155**, 63 (1988).
[36] C. Lebeau, A. Raboutou, P. Peyral, and J. Rosenblatt, Physica B **152**, 100 (1988).
[37] Z.Q. Wang and D. Stroud, Phys. Rev. B **44**, 9643 (1991).
[38] F.V. Kusmartsev, Phys. Rev. Lett. **69**, 2268 (1992).
[39] M.P.A. Fisher, T.A. Tokuyasu, and A.P. Young, Phys. Rev. Lett. **72**, 2931 (1991).
[40] H. Kawamura and M.S. Li, J. Phys. Soc. Jpn. **66**, 2110 (1997).
[41] A.R. Jurelo, I. Abrego-Castillo, J. Roa-Rojas, L.M. Ferreira, L. Ghivelder, P. Pureur, and P. Rodrigues, Jr., Physica C **311**, 133 (1999).
[42] L.I. Glazman and A.E. Koshelev, Phys. Rev. B **43**, 2835 (1991).
[43] X.G. Qiu, V.V. Moshchalkov, Y. Bruynseraede, and J. Karpinski, Phys. Rev. B **58**, 8826 (1998).