

## Field and oxygen dependence of the magnetic irreversibility line in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

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We report on dc magnetic susceptibility measurements in polycrystalline  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  as a function of the oxygen content in the limit of small  $\delta$ . We focus mainly on the detailed behavior of the zero-field-cooled–field-cooled irreversibility line at fields below 7 kOe. We find that this line shows a crossover from a  $H^2$  dependence at higher fields to the more commonly observed  $H^{2/3}$  behavior at lower fields. This feature is analogous to the anisotropy induced crossover from the high-field Gabay-Toulouse line to the low-field de Almeida-Thouless behavior, characteristic of the spin-glass transition. Our results are discussed in the light of a dirty-limit vortex-glass model where the grain charging energy plays a major role. Data concerning the behavior of the resistive transition as a function of the distribution of the oxygen vacancies are also presented.

### I. INTRODUCTION

The static and dynamic properties of vortices are among the most studied topics in the high temperature superconductors (HTSC). Real progress has been obtained in this field, since relatively simple phenomenological theories are useful for the understanding of several of the experimental observations.<sup>1,2</sup> The layered structure of these materials associated to strong type-II character are distinctive features underlying their vortex phenomenology. For most of the available samples also important either for statics or dynamics is the large distribution of defects, occurring in microscopic (e.g., oxygen vacancies), mesoscopic (e.g., twin boundaries or stacking faults), and macroscopic (voids, precipitates, or grain boundaries) scales.

Particularly concerning the collective vortex properties, detailed and extensive characterizations have been performed mainly related to experiments on flux lattice decoration,<sup>3</sup> neutron scattering,<sup>4</sup> and macroscopic magnetism.<sup>2</sup> Many of the observations are fairly well understood. However, some other properties remain unclear. One of the most interesting and widely studied of these topics is the so-called “irreversibility line,” which separates in the  $HT$  plane a magnetically irreversible region from a high-temperature regime, where the diamagnetic susceptibility is entirely reversible. This line runs neatly below  $H_{c2}(T)$ , and its origin is a matter of strong controversy. Two major interpretations have been put forward. One of them is based on the effects of thermally activated depinning of Abrikosov vortices. The theory<sup>5</sup> follows along the lines of the Anderson-Kim vortex thermodynamics<sup>6</sup> including flux-creep effects. However, the validity of this theory in the case of HTSC has been seriously questioned, since, in contrast to experiment, it predicts Ohmic  $V$ - $I$  behavior at any field and temperature, logarithmic decay of the trapped magnetization, and exponential decrease of the resistivity.

The alternative interpretation of the irreversibility line bases on the occurrence of a phase-transition

phenomenon.<sup>1,7</sup> This line could represent a melting frontier above which the vortices are in a fluid state and below which they are frozen into a lattice. Different models have been proposed. However, the similarities between the irreversibility line in HTSC and the one formerly observed in spin glasses<sup>8</sup> points towards a broken-ergodicity transition, where disorder and frustration are basic ingredients. According to this interpretation, just below the irreversibility line the system would be frozen in a vortex glass state<sup>7</sup> characterized by a highly degenerate but nontrivial ground state, constituted by many nonequivalent vortex configurations with approximately the same energy. Irreversibilities then result from limited excursions of the system in a complex many-valleyed phase space, as currently pictured for spin glasses and other frustrated and disordered systems.<sup>9</sup> A simple vortex glass model consisting of a collection of Josephson coupled grains was originally proposed<sup>10</sup> for granular superconductors. More recently this model and its generalizations have been applied with moderate success to explain the high-temperature dynamic properties of the vortex lattice in HTSC.<sup>11,12</sup> There is no theoretical proof, however, that such a glass phase exists below the irreversibility line, in spite of experiments<sup>13</sup> and theoretical<sup>11,12,14</sup> evidence.

In order to contribute with new elements to the subject of magnetic irreversibilities in HTSC, we report on an experimental study of the irreversibility line as a function of the oxygen concentration in a polycrystalline  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  sample. By varying the oxygen content of the same sample in the limit of low oxygen depletion, we are primarily concerned with the role of microscopic defects (the oxygen vacancies). The mesoscopic defect distribution may also be significantly altered by inclusions of metamorphic ortho-II or tetragonal phases. We show that the irreversibility line follows two different regimes in the low-field–high-temperature portion of the  $HT$  phase diagram. For  $H < 1$  kOe the irreversibility temperature depends on the field as  $H^{2/3}$ , the so-called “de Almeida–Thouless-like” line, while for higher fields a

crossover is observed to a reverted curvature  $H^2$  dependence. This behavior is entirely analogous to experimental results<sup>15,16</sup> and predictions of the mean-field theory<sup>17,18</sup> for the spin-glass transition. Based on these analogies, we propose a qualitative interpretation for the irreversibility line in deoxygenated  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  in terms of a vortex-glass transition.

## II. EXPERIMENTAL DETAILS

The samples were prepared from high quality  $\text{Y}_2\text{O}_3$ ,  $\text{BaCO}_3$ , and  $\text{CuO}$  by solid-state reaction. After being ground and sintered at  $950^\circ\text{C}$ , the samples were cooled slowly down to  $450^\circ\text{C}$  and annealed at this temperature for 3 days in oxygen atmosphere. Samples with densities ranging from 80 to 90 % of the ideal density were obtained. We consider these samples as being in a fully oxygenated state,  $\delta=0$  (most probably  $\delta\approx 0.05$ , as indicated by x-ray analysis). Subsequent deoxygenations were made in steps, by heating the samples in vacuum to  $300\text{--}450^\circ\text{C}$  for short times. The oxygen desorption was controlled by weight loss. Oxygen resorption was performed by heating the samples to  $450^\circ\text{C}$  in pure oxygen atmosphere. This oxygenation-deoxygenation technique gives reproducible results if contact of the sample with air is prevented. The simplicity and reproducibility of the process is an undoubted advantage of using ceramic samples in our study.

In addition to the reference state ( $\delta\approx 0$ ), we report data for  $\delta=0.09, 0.15, 0.16, 0.21$ , and  $0.26$ . For each oxygen content, the magnetic irreversibility line,  $T_{\text{irr}}(H)$ , and the electrical resistivity,  $\rho(T)$ , are studied. The irreversibility points were obtained from the splitting of the dc magnetic susceptibility measured in zero-field-cooled (ZFC) and field-cooled (FC) conditions. These measurements were performed with a vibrating sample magnetometer having a sensitivity better than  $10^{-4}$  emu in fields up to 7 kOe. The critical temperature determined from the maximum of the temperature derivative of the resistivity is approximately constant for all oxygen states. Deoxygenation basically enlarges the tail of the resistive transition. Most of the resistivity results will be discussed in a subsequent publication.

For conciseness, the results shown in this paper refer to the same specimen. Measurement on other samples showed entirely similar trends.

## III. RESULTS

Figure 1 shows typical dc-susceptibility curves for  $\text{YBa}_2\text{Cu}_3\text{O}_7$  measured under ZFC and FC conditions at two different fields. We observe that above a given temperature the ZFC and FC curves merge together, defining a perfectly reversible regime for the diamagnetic signal. In Fig. 2 are shown differences of the susceptibilities FC-ZFC in several fields, plotted as functions of the temperature for oxygen depletion  $\delta=0.16$ . The temperature  $T_{\text{irr}}(H)$ , where the difference between the FC and ZFC signals falls to zero defines the limit between irreversible and reversible regions. As the ZFC and FC curves join tangentially, our  $T_{\text{irr}}(H)$  are determined to an uncertain-

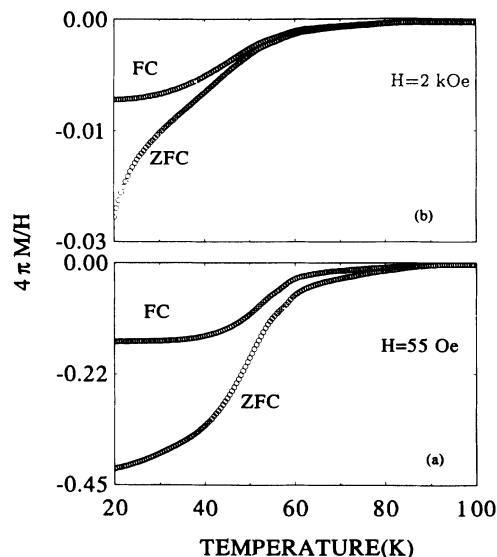


FIG. 1. Field-cooled (FC) and zero-field-cooled (ZFC) diamagnetic susceptibility of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  ( $\delta=0.26$ ) measured at (a) 55 Oe and (b) 2 kOe. The temperature  $T_{\text{irr}}(H)$ , corresponding to the splitting of the ZFC and FC curves, marks the onset of the irreversibility effects. Demagnetizing field is taken into account.

ty of  $\pm 0.5$  K in most cases. Otherwise, within the accuracy of our equipment, we have verified that  $T_{\text{irr}}(H)$  does not depend on the (slow) temperature drift during the experiments, at least up to a rate of 1 K/min, nor on the sense of the temperature variation for the FC measurements. We have also verified that the absolute values of

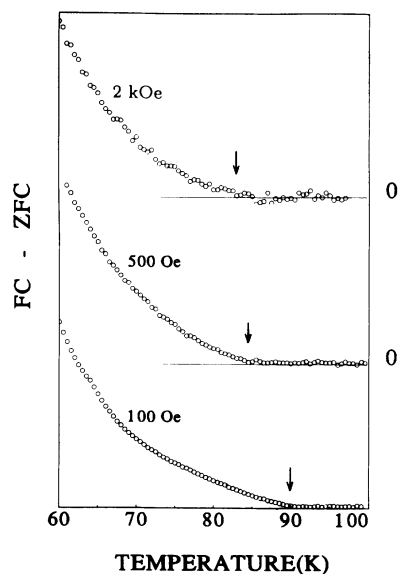


FIG. 2. Differences FC-ZFC in several field for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  ( $\delta=0.16$ ) plotted as functions of the temperature. The temperature  $T_{\text{irr}}(H)$ , where the difference between the FC and ZFC susceptibilities falls to zero, defines the limit between irreversible and reversible regimes. Arrows indicate  $T_{\text{irr}}(H)$ .

the dc susceptibility decrease substantially with oxygen depletion in the high-temperature region ( $T > 60$  K).

Figure 3 displays the  $T_{\text{irr}}(H)$  lines of a same specimen of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  for six different oxygen states, numbered in chronological order of experiments. Curve 1 is the irreversibility line for the reference state of the sample, for which we assume  $\delta \approx 0$ . Curves 2, 3, 4, 5, and 6 correspond respectively to  $\delta = 0.09, 0.15, 0.16, 0.26,$  and  $0.20$ . Curve 6 was obtained after partially reoxygenating from the state corresponding to  $\delta = 0.26$ . The qualitative shape of curves in Fig. 3 has been shown to be consistently reproducible for different samples of the HTSC.<sup>8,13,19–21</sup>

From the analysis of the data in Fig. 3, we obtain that in low fields all the  $T_{\text{irr}}(H)$  curves may be fitted to a de Almeida-Thouless<sup>22</sup> (AT) line

$$H^{2/3} = \alpha [1 - T_{\text{irr}}(H)/T_{\text{irr}}(0)], \quad (1)$$

where  $T_{\text{irr}}(0) = 93$  K and  $\alpha$  is a constant for a given oxygen content. Values for  $\alpha$  are shown in Table I. Above 1 kOe approximately, the experimental curves in Fig. 3 deviate strongly from the AT-like line. Moreover, from 2 kOe approximately and up to the highest applied fields ( $\sim 7$  kOe),  $T_{\text{irr}}(H)$  for curves 3, 4, 5, and 6 fit well to a reverted curvature  $H^2$  line:

$$H^2 = \beta [1 - T_{\text{irr}}(H)/T_{\text{irr}}(0)], \quad (2)$$

where  $T_{\text{irr}}(0) = 83$  K and  $\beta$  is tabulated in Table I. Note that the high-field behavior of curves 1 and 2 is also compatible with Eq. (2), although it does not allow the determination of  $\beta$ . We call the behavior represented by Eq. (2) a Gabay-Toulouse-like line, in analogy to the Gabay-Toulouse<sup>23</sup> (GT) transition of the spin-glass systems.

The crossover observed in the irreversibility lines of

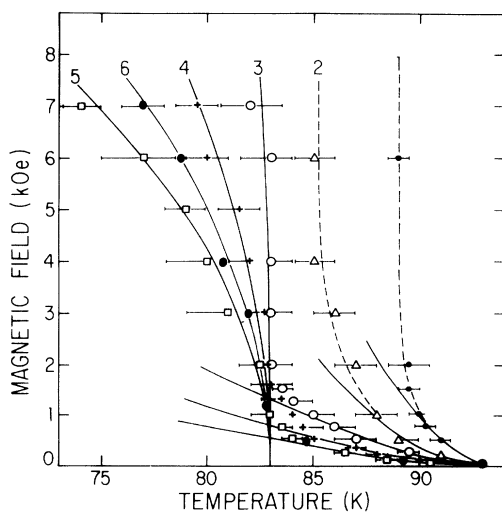


FIG. 3. Irreversibility lines for a given specimen of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  for six different oxygen states. Curves 1, 2, 3, 4, 5, and 6 correspond, respectively, to  $\delta \approx 0.0, 0.09, 0.15, 0.16, 0.26,$  and  $0.20$ . Continuous lines with positive and negative curvatures correspond to fits to Eqs. (1) and (2), respectively. Fitting parameters are given in Table I.

TABLE I. Parameters obtained from fits of the experimental data in Fig. 3 to Eqs. (1) and (2). The reference temperatures for the de Almeida-Thouless-like and Gabay-Toulouse-like expressions are  $T_{\text{irr}}^{\text{AT}}(0) = 93$  K and  $T_{\text{irr}}^{\text{GT}}(0) = 83$  K, respectively.

Oxygen depletion ( $\delta$ )	$\alpha$ (kOe) <sup>2/3</sup>	$\beta$ (kOe) <sup>2</sup>
0.0	30.7	
0.09	19.5	
0.15	11.0	
0.16	8.0	1200
0.20	7.5	690
0.26	6.0	500

Fig. 3 from the high-field  $H^2$  regime to the low-field  $H^{2/3}$  regime is reminiscent of the transition of a weakly anisotropic spin-glass system. Indeed, the mean-field theory for Heisenberg or  $XY$  spin glasses predicts that when the reduced field is small enough compared to the ratio anisotropy/exchange, the transition becomes Ising-like.<sup>17,18</sup> Experimentally, one observes a crossover in the critical irreversibility line from the Gabay-Toulouse to the de Almeida-Thouless dependence in the limit of low applied fields.<sup>15,16</sup>

Concerning the remarkable  $H^2$ - $H^{2/3}$  crossover, Fig. 3 reveals some features that are reproducible for different samples. The functional form of  $T_{\text{irr}}(H)$  does not depend on the deoxygenation, but  $\delta$  systematically affects the coefficients  $\alpha$  and  $\beta$  in Eqs. (1) and (2) (see Table I). The crossover is broad for small  $\delta$  but becomes sharper and moves to lower temperatures for increasing  $\delta$ . For the curves corresponding to  $\delta \geq 0.15$ , the extrapolated  $T_{\text{irr}}(0)$  in Eq. (2) are coincident to 83 K.

The relation between curves 3 and 4 in Fig. 3 also deserves some comments. Curve 3 corresponds to  $\delta = 0.15$ . This oxygen state was obtained from the precedent  $\delta = 0.09$  by means of a rapid deoxygenation process (10 min at  $T = 400^\circ\text{C}$ , in vacuum). Presumably, an inhomogeneous distribution of oxygen vacancies is produced, as depletion is supposed to proceed at first from regions close to grain boundaries, voids, and other surface defects. Subsequent to the magnetic and resistivity measurements, the sample was submitted to an annealing in vacuum at  $400^\circ\text{C}$ , for 80 h. The average oxygen content hardly changes. We determined  $\delta = 0.16$  for the oxygen depletion in curve 4. Consequently, the noticeable changes in coefficients  $\alpha$  and  $\beta$  from curves 3 to 4 are due basically to a rearrangement of the oxygen vacancies: the inhomogeneous distribution of curve 3 evolves to an homogeneous state in curve 4. This rearrangement of the vacancies is clearly confirmed by the resistive transition shown in Fig. 4. There one observes that for the quickly deoxygenated  $\delta = 0.15$  sample, the resistive regime describing the approach to the zero resistance state presents the inverted curvature typical of a percolation transition. For  $T < 82$  K, the resistivity may be fitted to a power law of the type

$$\rho \sim (T - T_{c0})^s, \quad (3)$$

where  $s = 0.80 \pm 0.05$  and  $T_{c0} = 76.0$  K. This exponent is consistent with theoretical calculations for three-

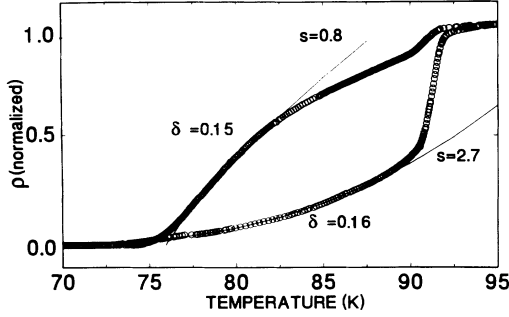


FIG. 4. Resistive transition at zero applied field for the oxygen depletions  $\delta=0.15$  and  $\delta=0.16$ , corresponding to curves 3 and 4, respectively, in Fig. 3. Continuous lines are fits to Eq. (3) with the quoted exponents.

dimensional (3D) percolation<sup>24</sup> and has been observed in granular thin films prepared by coevaporation of metal-insulator mixtures.<sup>25</sup> Thus, our inhomogeneous  $\delta=0.15$  sample behaves as a 3D assembly of weakly coupled superconducting grains, where barriers for pair propagation are produced by strong deoxygenation of the crystallite frontiers.

On the other hand, for the homogeneous state  $\delta=0.16$ , the resistivity in the  $R \rightarrow 0$  regime is significantly lower and presents a long tail characteristic of thermal fluctuations. Indeed, as shown in Fig. 4, between 78 and 88 K the resistivity can be fitted to Eq. (3) with the exponent  $s = 2.7$ . Below 80 K down to  $T_{c0} \cong 73$  K, a best fit is obtained with  $s = 1.5$ . The exponent  $s = 2.7$  has been found in polycrystalline HTSC and other artificially structured granular superconductors,<sup>26</sup> and is interpreted as the critical exponent for the paracoherent region ( $T > T_{c0}$ ) of a intergranular coherence transition.<sup>27</sup> The exponent  $s = 1.5$  has been observed in several high- $T_c$  ceramics<sup>28</sup> and is also ascribed to thermal fluctuations. The prevailing thermodynamic effects over geometrical percolation in the resistive transition indicate that for the  $\delta=0.16$  case, inhomogeneities underlying the system behavior are operating in a range scale of the order of the coherence length. This suggests a more uniform distribution of oxygen vacancies, as compared to the former  $\delta=0.15$  case.

#### IV. DISCUSSION

It is generally thought that a good description of the granular HTSC is given in terms of a 3D Josephson medium. Such a model should be acceptable even for defect-enhanced and twinned single crystals. The large distribution of defects characteristic of most HTSC samples gives rise to short-range spatial modulations of the superconducting order parameter.<sup>29</sup> Owing to the relatively larger penetration depth, it is hardly conceivable that the mixed state of these systems would be characterized by a regular array of homogeneous Abrikosov tubes. In most cases, screening currents flowing around a given vortex core should cross a number of defective areas and Josephson barriers are likely to be present.

A simple representation of the inhomogeneous system is given by a 3D network of weakly coupled grains de-

scribed by the following effective Hamiltonian:

$$H = -2e^2 \sum_{i,j} n_i n_j C_{i,j}^{-1} - \sum_{i,j} J_{ij} \cos(\theta_i - \theta_j - A_{ij}), \quad (4)$$

where  $n_i$  and  $\theta_j$  are canonically conjugate variables,<sup>30</sup>

$$[n_i, e^{i\theta_j}] = \delta_{ij} e^{i\theta_j}, \quad (5)$$

representing, respectively, the number of pairs on grain  $i$  and the phase of the Ginzburg-Landau order parameter, supposed uniform inside grain  $j$ . In Eq. (4)  $C_{ij}^{-1}$  is the inverse capacitance matrix,  $J_{ij}$  is the coupling energy between grains  $i$  and  $j$  and

$$A_{ij} = \frac{2\pi}{\phi_0} \int_i^j \mathbf{A} \cdot d\mathbf{l}, \quad (6)$$

where  $\phi_0$  is the elementary flux quantum,  $\mathbf{A}$  is the vector potential, and the limits of the line integral are the centers of grains  $i$  and  $j$ . The first term of the Hamiltonian, Eq. (4), represents a Coulomb energy, while the second term is a lattice generalization of the Josephson coupling between two superconducting grains.

The possibility of occurrence of frustration is an interesting feature of the model defined by Eq. (4). When a magnetic field is applied, frustration is induced when the random phase factors  $A_{ij}$  are distributed over a wide range compared to  $2\pi$ . Moreover, in a process discussed by Spivak and Kivelson,<sup>31</sup> the Josephson energies  $J_{ij}$  may change sign to  $-J_{ij}$  if there is an impurity state between grains  $i$  and  $j$  being occupied by one electron with spin up (down). In disordered materials containing  $\text{Cu}^{2+}$  ions as the HTSC, one may expect that such negative Josephson couplings occur commonly. The resulting  $\pm J$  random distribution may produce frustration even in the limit of vanishing small magnetic fields.

Using the pseudospin representation,<sup>27</sup>  $S_i^\pm = \exp(\pm i\theta_i)$ , considering only one site Coulomb interactions<sup>32</sup> and following along the same lines as Lebeau *et al.*,<sup>33</sup> the classical limit of Eq. (4) may be written as

$$H = - \sum_{ij} \tilde{J}_{ij} S_i^+ S_j^- - \sum_i D_{ii} S_{ii}^2, \quad (7)$$

where

$$\tilde{J}_{ij} = J_{ij} \cos A_{ij} \quad (8)$$

and

$$D_{ii} = \frac{2e^2}{C_{ii}}. \quad (9)$$

The  $C_{ii}$  are the capacity coefficients of isolated grains.<sup>34</sup> The effective exchange  $\tilde{J}_{ij}$  are randomly distributed with mean value equal to zero. Randomness comes from disorder, applied field, and negative local superfluid densities, which invert the sign of  $J_{ij}$ . In Eq. (7) we drop antisymmetric terms<sup>35</sup> whose amplitudes,  $J_{ij} \sin A_{ij}$ , are supposed to be small in the limit of small fields and moderate disorder.

The model represented by Eq. (7) is formally analogous to the vector  $XY$  spin glass Hamiltonian with random uniaxial anisotropy. The coefficients  $D_{ii}$  play the role of a

weak and local anisotropy strength. Kotliar and Sompolinsky<sup>17</sup> and Fischer<sup>18</sup> treated theoretically a similar problem for a Sherrington-Kirkpatrick spin glass with Heisenberg spins. They showed that the spin-glass transition presents a crossover in the  $HT$  plane from the high-field Gabay-Toulouse<sup>23</sup> dependence to an Ising-like behavior (the de Almeida-Thouless line<sup>22</sup>) in low fields, the strength of the field being defined with respect to the ratio  $D/J$ . This kind of crossover has been experimentally identified in the irreversibility line of spin glasses.<sup>15,16</sup>

Since the pioneering work of Shih, Ebner, and Stroud,<sup>10</sup> many theoretical studies are available on equilibrium and nonequilibrium properties of the model vortex-glass represented by Eq. (4). Most of them, however, do not take into account the charge energy term. Specifically, in this model the irreversibility line is interpreted as dividing the  $HT$  phase diagram into ergodic and nonergodic states.<sup>36</sup> The predictions concerning the shape of this line at small fields are not clear. Numerical studies on 2D and 3D small arrays of point grains<sup>10</sup> indicate an Ising-like transition. However, the  $H^{2/3}$ -AT behavior, as shown by Morgenstern, Müller, and Bednorz,<sup>37</sup> would be somewhat accidental and dependent on a site-type disorder where grains are weakly displaced from their positions in the quiescent square lattice. A more recent analytical calculation by Sergeenkov,<sup>38</sup> based on the model vortex-glass Eq. (4), where the charge energy term is neglected, gives an irreversibility line of the  $H^2$ -Gabay-Toulouse-type. None of these theories, however, is able to explain the GT-AT crossover observed in Fig. 3. An earlier explanation proposed by Sergeenkov,<sup>39</sup> based on the effect of field-induced dynamics of dislocation-mediated weak links, indeed predicts a crossover but from a low-field  $H^{2/3}$ -AT-like behavior to a high-field  $H^q$  dependence, where the exponent  $q$  varies continuously under deoxygenation from  $q=2$  to  $\frac{1}{4}$ . This is in contradiction with our results, where both the GT and AT exponents are independent of the oxygen deficiency.

Recently, efforts have been made to introduce the charge energy term of Eq. (4) in the model calculations. This contribution becomes relevant in the limit of small grains,<sup>40</sup> which seems to be the case of the HTSC. Transport properties,<sup>41</sup> including  $I$ - $V$  characteristics, and the interesting possibility for a new orbital glass state<sup>42</sup> have been theoretically investigated in connection to Eq. (4). To our knowledge, however, there is no calculation of the irreversibility line starting from the full Hamiltonian, Eq. (4). Based on the theoretical and experimental analogies between the spin glass and the granular superconductor problems, we are thus led to suggest that a phenomenon equivalent to the GT-AT crossover is likely to explain the shape of the irreversibility lines shown in Fig. 3. The close resemblance of the spin-glass transition calculated by Fischer,<sup>18</sup> or the experimental data by Kenning, Chu, and Orbach<sup>16</sup> on Cu-Mn, and the irreversibility lines for our deoxygenated granular  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is indeed remarkable.

In the spin-glass theory, the reference freezing temperature  $T_f^{\text{GT}}(0)$  for the GT line differs from the characteristic temperature  $T_f^{\text{AT}}(0)$  for the AT line. For  $XY$  spin

glasses, the theory of Fischer<sup>18</sup> predicts that

$$T_f^{\text{GT}}(0) - T_f^{\text{AT}}(0) = -\frac{2}{k_B\sqrt{3}}D. \quad (10)$$

Identifying our extrapolated zero-field irreversibility temperatures  $T_{\text{irr}}^{\text{GT}}(0)$  and  $T_{\text{irr}}^{\text{AT}}(0)$  with the spin-glass freezing temperatures in Eq. (10), we estimate that the anisotropy strength varies between 0.3 to 0.7 meV. This allows us to calculate the capacity coefficients from Eq. (9). On the other hand, from the expression for the charge energy given by Abeles *et al.*<sup>43</sup> and supposing that superconducting and insulating grains have about the same average size, we obtain that  $C \sim \epsilon d$ , where  $\epsilon$  is the dielectric constant and  $d$  is the typical grain size. Therefore, using the capacity coefficients calculated from Eqs. (10) and (9) and the average dielectric constant  $\epsilon \approx 300$ , as given by Behrooz and Zettl,<sup>44</sup> we obtain that  $d$  ranges between 0.1 and 0.3  $\mu$ . Given the several approximations involved, this should be considered an order of magnitude estimation. Nevertheless, this value for  $d$  is in agreement with other experimental estimations,<sup>8,13</sup> including microwave absorption at low magnetic fields.<sup>45</sup> Moreover, the estimated  $d$  has the order of magnitude of the London penetration depth, consistently with assumptions underlying the vortex-glass Hamiltonian, Eq. (4).

From Fig. 3 and Table I, we observe that the coefficient  $\alpha$  of Eq. (1) decreases regularly upon deoxygenation. Particularly concerning this coefficient, the simplest superconducting glass models based on Eq. (4) (Refs. 10 and 35) predict that  $\alpha$  is proportional to the ratio  $\phi_0/S$ , where  $S$  is the mean projected area of closed loops of superconducting grains. According to these theories, a decrease of  $\alpha$  implies an increase of  $S$ . This might be expected if a number of junctions are weakened or destroyed in the deoxygenation process. In this case, the length of the average percolation loop, which binds the area  $S$ , would be effectively increased. It is also clear from Fig. 3 and Table I that the coefficient  $\beta$  of the GT-like line, Eq. (2), decreases when the sample is deoxygenated. This suggests that  $\beta$  is also dependent on the geometrical characteristics of the granular array.

The above outlined interpretation, based on the spin-glass phenomenology, in describing the behavior of the irreversibility line in deoxygenated  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , is subject to many criticisms. One of them concerns of course the validity of the close analogy proposed between the  $XY$ -spin glass and the superconducting vortex glass problems. It is clear that the two problems are not identical. For instance, the effective Hamiltonian Eq. (4) lacks time-reversal symmetry in contrast to its spin counterpart. Going further along this line, one may wonder about the very existence of a glass transition at finite temperatures in a system of  $XY$  spins. There are evidences that the short-range vector-spin models do not show a conventional spin-glass transition in spatial dimension  $d \leq 4$ .<sup>46</sup> In this respect, however, the situation is far from being clear. The experiments are rather consistent with the phase transition scenario at nonzero temperatures.<sup>47</sup> Moreover, it was recently found theoretically that the anisotropy plays a crucial role by inducing the phase transition at nonzero  $T$ .<sup>48</sup> Specifically concerning the dirty-

limit vortex-glass superconductor modeled by Eq. (4), results from Monte Carlo simulation<sup>14</sup> also suggest the occurrence of a spin-glass-like ordered phase at finite temperatures. However, this contrasts with real-space renormalization-group studies,<sup>35</sup> which indicate that the lower critical dimension for a vortex-glass transition is greater than 3.

On the other hand, it is difficult to understand the overall behavior of the irreversibility lines in Fig. 3 within the flux-creep picture. It was proposed that the AT-like line results from a giant flux-creep effect.<sup>5</sup> The same mechanism can hardly be conceived to produce the crossover to the GT-like behavior.

The irreversibility line can also result from a more conventional flux-lattice melting induced by thermal fluctuations. Using a Lindemann-type criterion for melting of the vortex lattice in an anisotropic 3D medium, Houghton, Pelcovits, and Sudbo<sup>49</sup> found that at low field the transition is approached as  $H^{1/2}$ . Our data are better described by the  $H^{2/3}$  dependence, and no crossover of type shown in Fig. 3 is predicted by these authors. Recently, Glazman and Koshelev<sup>50</sup> proposed that at high fields both fluctuations and Lindemann melting of the vortex-lattice become effectively two dimensional. In this case, a crossover in the melting line would be expected. However, in the high-field region the authors<sup>50</sup> predict that the melting line is essentially field independent. This is in contradiction with the inverted  $H^2$  curvature clearly demonstrated by the results in Fig. 3.

## V. CONCLUSION

We measure the dc magnetic susceptibility and the resistivity of a polycrystalline sample of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  as a function of the oxygen content. We restricted ourselves to low-oxygen depletions. By performing ZFC and FC experiments, we are able to study in detail the magnetic irreversibility line for different oxygen states in the low-field limit. The irreversibility temperatures clearly show a progressive change from a  $H^2$  dependence at fields larger than  $\sim 1$  kOe to the more commonly observed  $H^{2/3}$  behavior at smaller fields. This crossover and the  $H^2$  line are more clearly defined for the deoxygenated

states of the sample. We suggest an interpretation for these observations, which takes into account the analogies presented by the spin glasses and the inhomogeneous superconductors. Our description is based on a dirty-limit vortex glass model, which is supposed to be valid when the superconductor behaves as a granular Josephson medium. Within this scheme, the irreversibility line is due to a broken ergodicity transition where the basic ingredients are disorder and frustration. The detailed field dependence of the irreversibility line is proposed to be the vortex-glass analog of the effective dimensional crossover from the high-field Gabay-Toulouse transition to the low-field de Almeida-Thouless instability observed in spin glasses. In the superconductor, the charging energy plays the role of the anisotropic interaction inducing the crossover.

The spin-glass-vortex-glass analogy also allows us to estimate that the regions of homogeneous phase have an average size of the order of  $0.1 \mu$ , which is much smaller than the typical metallurgical grains. This implies that superconducting granularity in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is a subtle concept, which is not trivially related to polycrystallinity. Indeed, our results concerning the irreversibility line and the resistive transition indicate that granularity in this system is strongly associated with the distribution of oxygen vacancies. Most probably, the vortex glass state in polycrystalline  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  is tributary of the whole distribution of defects, including mesoscopic and macroscopic scale defects in addition to the microscopic ones.

As a general conclusion, our experiments in the low-field-high-temperature portion of the  $HT$  phase diagram of the polycrystalline high- $T_c$   $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  superconductor, give significant evidence for a broken-ergodicity transition separating a higher temperature and reversible fluidlike vortex phase from a dirty-limit vortex glass state at lower temperatures.

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