# Glass behavior and the H-T phase diagram of the high- $T_c$ ceramic superconductors YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, and GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>

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We report systematic field-cooled (FC) and zero-field-cooled (ZFC) diamagnetic susceptibility measurements in polycrystalline samples of the high- $T_c$  oxide superconductors  $R\text{Ba}_2\text{Cu}_3\text{O}_7$ , where R=Y, Eu, or Gd. We focus mainly on the temperature range near  $T_c$ , where the FC and ZFC curves split up below a characteristic temperature, similar to the behavior of spin glasses. This temperature depends on the applied magnetic field and defines a crossover line in the H-T plane separating ergodic and nonergodic behavior, in close resemblance to what is found in conventional spin glasses. We suggest that this is a general characteristic of the ceramic oxide superconductors. Moreover, our data allow us to obtain a more detailed high-temperature H-T phase diagram, where a lower temperature bound for the superconducting glasslike state is proposed.

### I. INTRODUCTION

There is now much experimental and theoretical evidence for the existence of a close relationship between the nature of the superconducting state in oxide superconductors and inhomogeneities occurring in these materials. The picture that emerges differs qualitatively in many respects from the conventional Abrikosov state, at least at temperatures close to  $T_c$ . Müller, Takashige, and Bednorz1 were the first to propose the existence of a superconducting glass state in the La-Ba-Cu-O system, based on their measurements of magnetic properties reminiscent of the behavior of spin glasses. Since then, many experimental results concerning magnetic hysteresis,2 diamagnetic susceptibility,3,4 magnetic relaxation,5,6 electrical resistivity, and the temperature derivative of the resistivity, magnetoresistance, low-field microwave absorption, critical field and critical current density, 7,11 among others, clearly point to an inhomogeneous and multiconnected superconducting structure for describing the new oxide systems, as well as the earlier low- $T_c$  system  $\mathrm{BaPb}_{1-x}\mathrm{Bi}_x\mathrm{O}_3$ . The generally accepted picture is that of a disordered network of superconducting grains, where the overall phase coherence is established through weak couplings between the grains. Such a granular model is particularly appealing for the ceramic samples, owing to their porosity and the possible insulating on normal character of the crystallite surfaces, induced by a depletion of oxygen content and/or by a supression of superconductivity on grain boundaries due to a short coherence length.<sup>13</sup> An inhomogeneous superconducting picture has also been proposed for single-crystal samples on the basis of the observed high density of extended defects such as twin boundaries and stacking faults, or to variable oxygen stoichiometry.<sup>13</sup>

The granular model may be only an approximate description of an actually more complex fractal cluster structure, but it has the advantage of having been theoretically investigated to a large extent. 14,15 One of

the most interesting features predicted by the theory of granular superconductors is the possibility of an occurrence of field-induced frustration, which together with disorder leads to "glassy" properties that are analogous to those of conventional spin glasses.<sup>14</sup>

In this work we report detailed magnetic-susceptibility measurements in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, and GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> polycrystalline samples. Our results reveal spin-glass-like irreversibilities and allow for the determination of a phase diagram in the field-temperature plane. This *H-T* phase diagram seems to be a general, sample independent, characteristic of the superconducting oxides and also depicts close resemblances to the corresponding one for spin-glass systems. <sup>16</sup>

## II. EXPERIMENT

Our polycrystalline samples have been prepared by the usual solid-state reaction technique<sup>17</sup> under flowing oxygen atmosphere in a Pt crucible for 6 h. After powdering the samples, they were pressed into discs of about 2 mm in thickness and 9 mm in diameter, and sintered at 950 °C for 16 h and then cooled slowly, especially between 800 °C and 500 °C, down to room temperature. X rays have shown the expected orthorhombic phase. The densities of the YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, and GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> samples are respectively 5.33, 5.99, and 5.286 g/cm<sup>3</sup>.

The magnetic measurements were performed with a Foner magnetometer, having a sensitivity of  $10^{-4}$  emu, in fields up to 8 kOe. The temperature was read by means of a carbon-glass sensor. In order to minimize the demagnetizing fields, the samples were held with their diameter parallel to the applied field.

## III. RESULTS

Figure 1 exemplifies typical dc-susceptibility results as a function of temperature in two different applied fields for the GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> system. Similar data were obtained for the three systems studied in several applied fields. In

order to obtain a meaningful interpretation, all measurements were performed in strict observance of an invariable protocol. In a given run the sample was cooled down to 4–10 K, either in zero applied field or in a constant field. The data were then recorded while warming up the sample at a constant rate of approximately 0.5 K/min.

The field-cooled (FC) curves are practically reproducible, either for cooling down or heating up the sample, and are assumed to represent close-to-equilibrium states of the system. \(^{14}\) On the contrary, the zero-field-cooled (ZFC) curves are strongly irreversible with respect to thermal cycling. Moreover, the magnitude of the ZFC susceptibility decays with time appreciably when the temperature is held fixed. It is worth noticing in Fig. 1 the existence of a totally reversible behavior close to  $T_c$  (defined as the onset of the diamagnetic contribution to the susceptibility). A well-defined temperature  $T_g(H)$  marks the ZFC and FC bifurcation, thus separating the reversible and nonreversible regimes.

Results such as those in Fig. 1 are often taken as indi-

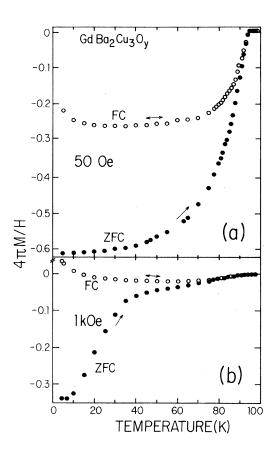


FIG. 1. Field-cooled (FC) and zero-field-cooled (ZFC) diamagnetic susceptibilities of polycrystalline  $GdBa_2Cu_3O_7$  measured at (a) 50 Oe and (b) 1000 Oe. The temperature  $T_g$  corresponding to the splitting of the ZFC and FC curves marks the onset of the irreversibility effects on the magnetic properties. Demagnetizing field is taken into account.

cative of the occurrence of a superconducting glass state in the oxide superconductors, because of their evident analogies with the susceptibility of spin-glass systems. According to this interpretation,  $T_g(H)$  is the analog of the glass temperature and denotes the limit between ergodic and nonergodic behaviors in the sense currently admitted for spin glasses. 18 It has been argued that such ZFC-FC irreversibilities may also result from a strongly enhanced flux creep effect, and this is proposed to be the case in single-crystal samples. 19 However, one should note that the intrinsically short coherence length together with evidences of internal Josephson tunnelling and the occurrence of irreversibilities even for fields below  $H_{c1}$ lend support to an inhomogeneous and frustrated picture for describing superconductivity in ceramic samples, at least at temperature approaching  $T_c$ .

Figures 2 and 3 give an extended view of the effects caused by different fields on the Y-, Eu-, and Gd-based samples, in the FC and ZFC conditions. When the field is increased, the susceptibility is strongly depressed. This effect is particularly pronounced in the ZFC curves when the temperature approaches  $T_c$ . An interesting aspect to be noted in Figs. 3(a) and 3(b) is the strong-field dependence of the point  $T_M(H)$ , where the Meissner (FC) signal departs from its low-temperature saturating behavior. For the Gd-based system (Fig. 2), a paramagnetic contribution raises the susceptibility towards positive values at low temperatures. The magnetic moment per Gd ion, as determined from a Curie-Weiss term above  $T_c$ , is almost equal to the free-ion value, in accordance with previous determinations. For the Eu-based compound we detect

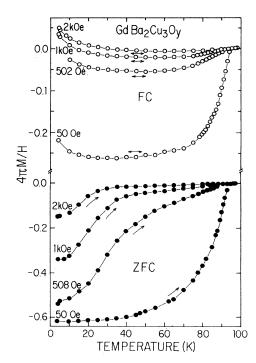


FIG. 2. Separate FC and ZFC susceptibilities for the GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> system in several applied fields.

TABLE I. Parameters obtained from fits of the low-field experimental data in Fig. 4 to the de Almeida-Thouless-type expression, Eq. (1).  $T_c(0)$  is taken as the temperature where the susceptibility first shows a decrease due to the superconducting diamagnetic contribution.

Compound	$a (kOe)^{2/3}$	T <sub>g</sub> (O) (K)	$T_c(O)$ (K)
YBa <sub>2</sub> Cu <sub>3</sub> O <sub>7</sub>	19.8	92.5	93
EuBa <sub>2</sub> Cu <sub>3</sub> O <sub>7</sub>	10.1	93.5	93.5
GdBa <sub>2</sub> Cu <sub>3</sub> O <sub>7</sub>	8.3	95	95

a paramagnetic susceptibility above  $T_c$  which does not follow a Curie-Weiss law. This result was expected since the Eu ion is known to behave nearly as in the free-ion case, giving a nonmagnetic ground state at low temperatures. For the Y-based system we also measure a positive susceptibility above  $T_c$ , which may be roughly fitted to a constant plus a Curie-Weiss term.

In order to study the ergodicity breaking phenomena in our samples, we perform very careful susceptibility measurements in the temperature range near  $T_c$ . These experiments allow us to determine  $T_g(H)$  with an accuracy limited mainly by the fact that the ZFC and FC

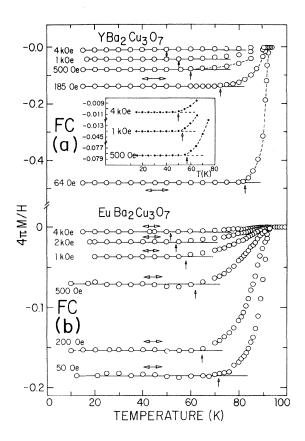


FIG. 3. FC susceptibilities for the (a)  $YBa_2Cu_3O_7$  and (b)  $EuBa_2Cu_3O_7$  systems. The arrows indicate the upper limit,  $T_M$ , of the temperature-independent behavior of the Meissner (FC) susceptibility.

branches join tangentially. Figure 4 shows results obtained for  $EuBa_2Cu_3O_7$ . The characteristic temperatures  $T_g(H)$  define lines in the H-T plane which look quite similar for the three studied systems, as shown in Fig. 5. These results are also similar to those previously reported for the La-Ba-Cu-O (Ref. 1) and Y-Ba-Cu-O (Ref. 4) systems. Recent measurements on a Bi-Sr-Ca-Cu-O superconductor reveal the same qualitative behavior. All of these results seem to point to a universal characteristic behavior of the polycrystalline oxide superconductors.

In spin glasses and other glasslike systems, the glass temperature, which denotes the limit between reversible and nonreversible regimes, is found to follow a line given by

$$H^{2/3} = a[1 - T_g(H)/T_g(0)],$$
 (1)

where  $T_g(H)$  corresponds to the onset of the irreversibilities under a field H,  $T_g(0)$  is the glass temperature at zero field, and a is a constant. The so-called de Almeida-Thouless (AT) line [Eq. (1)] was first derived from a mean-field theory for Ising spins. For low enough values of the magnetic field, our  $T_g(H)$  data also fit to a de Almeida-Thouless-type line as shown by solid curves in Fig. 5. The parameters of the best fits to Eq. (1) for the three systems studied are shown in Table I. We note that the parameter a, as well as the general shape of the curve  $T_g(H)$  for the Y-based system, are in good agreement with those previously reported for a sample of the same system but having much lower mass den-

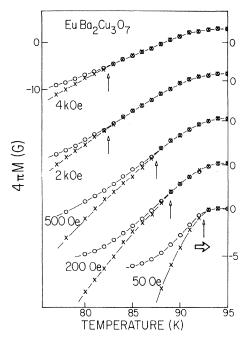


FIG. 4. Detailed FC (open circles) and ZFC (crosses) magnetizations for EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> in the range of temperatures near  $T_c$ . Measuring fields are quoted. Vertical arrows indicate  $T_g(H)$ , the temperature which signals the splitting of the FC and ZFC branches.

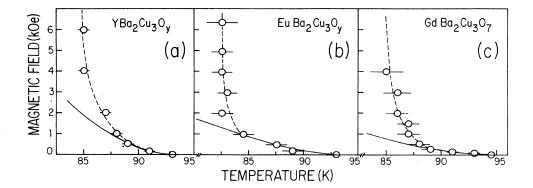


FIG. 5. Experimentally determined  $T_g(H)$  points as in Fig. 4, but for the three studied compounds. For each system the points define a line which separates ergodic and nonergodic regimes in the H-T phase diagram. The solid lines correspond to fits with the de Almeida-Thouless-type expression, Eq. (1), with coefficients given in Table I. Dashed lines are guides for the eye.

sity and having an order-of-magnitude higher room-temperature resistivity. Table I also shows that  $T_g$  tends to coincide with  $T_c$  in the limit of vanishing fields.

### IV. DISCUSSION

The Hamiltonian commonly proposed to describe a weakly coupled array of superconducting grains is written as 13

$$\mathcal{H} = -\sum_{i,j} J_{ij}(r_{ij}, \boldsymbol{B}, T) \cos(\theta_i - \theta_j - \boldsymbol{A}_{ij}) . \tag{2}$$

Here  $J_{ij}$  is the coupling energy between grains i and j (which depends on the intergrain distance,  $r_{ij}$ , the magnetic field B, and the temperature),  $\theta_i$  is the phase of the order parameter in grain i, and

$$A_{ij} = \frac{2\pi}{\phi_0} \int_i^j \mathbf{A} \cdot d\mathbf{l} \ , \tag{3}$$

where  $\phi_0$  is the elementary flux quantum, A is the vector potential, and the limits of the line integral are the centers of grains i and j.

The form of  $J_{ij}$  depends essentially on the nature of the couplings, which may be due to Josephson effect or to proximity effect. <sup>14</sup> In many applications of Eq. (2),  $J_{ij}$  is taken as a single positive constant for nearest-neighbor grains and zero otherwise. The possibility of occurrence of frustration when a magnetic field is applied is the most interesting feature of the model defined by Eqs. (2) and (3). Following Ref. 23, the Hamiltonian (2) may be rewritten as

$$\mathcal{H} = -\operatorname{Re} \sum_{i,j} K_{ij} S_i^* S_j , \qquad (4)$$

which shows that the system is isomorphic to a classical XY model for pseudospins  $S_i = \exp(i\theta_i)$ , where frustration is introduced by the phase factor  $A_{ij}$  in the effective couplings  $K_{ij} = J_{ij} \exp(iA_{ij})$ . Indeed, for sufficiently large values of B, the  $A_{ij}$  may be distributed over a wide range compared to  $2\pi$ . The model predicts the possible occurrence of a superconducting phase glass state where the

overall configuration of the order parameter consists of a random distribution of locally frozen phases. In fact, frustration and disorder lead the system into a situation of energetically nearly degenerate configurations close to the ground state. This is known to result in nonergodic glasslike behavior below the transition temperature. <sup>18</sup>

Some of the equilibrium and nonequilibrium properties of the model represented by Eqs. (2) and (3) have been investigated by a number of authors. Shih, Ebner, and Stroud<sup>14</sup> have performed numerical studies on twodimensional (2D) and three-dimensional (3D) ordered and disordered arrays of superconducting grains. Although their calculations are limited to relatively small arrays, many of the obtained properties are expected to be valid in the limit of large systems. In particular, in 3D disordered arrays they calculated a field-dependent transition temperature to a "phase glass" state, which looks qualitatively very similar to the experimental results shown in Fig. 5. In the low-field regime, the transition temperature decreases rapidly with increasing fields, but when frustration saturates the transition temperature line flattens and becomes nearly independent of the field, giving a picture quite analogous to Fig. 5. Numerical simulations were also performed by Morgenstern et al.,24 who generalize the granular 2D model by allowing for decoupled superconducting clusters inside physical grains. When they consider weak site disorder in a quiescent square lattice, and not too high values of the applied field, they find that an AT-like line quite closely describes the separation between equilibrium and metastable behavior in the susceptibility. The same quasi-de Almeida-Thouless transition was obtained by Schneider et al.25 who developed a mean-field analysis extending Abrikosov's theory of homogeneous type-II superconductors to inhomogeneous materials. An interesting analytical calculation was recently presented by Aksenov and Sergeenkov.<sup>23</sup> They propose a formulation based on Eq. (4) and consider site disorder over grain coordinates in a 2D lattice, obtaining a phase diagram in the H-T plane for the superconducting glass phase transition showing three different regimes. When the field is weak, typically corresponding to less

than one quantum of flux per plaquette, there is no frustration and the theory predicts the occurrence of a Meissner-type state. For intermediate fields, in the interval  $\frac{3}{4}H_0 < H < 15H_0$ , where the characteristic field is  $H_0 = \phi_0/2S$  and S is the mean plaquette area, a transition from normal to a phase glass state is predicted, following an AT-like line in the form

$$H^{2/3} = \left[\frac{3^{1/2}H_0^2}{2}\right]^{1/3} \left[1 - T_c(H)/T_c(0)\right]. \tag{5}$$

For higher values of the field, strong frustration drives the system into a true spin-glass-like phase with magnetization going to zero and  $T_c$  no longer depending on the field, as previously found in numerical calculations by Shih, Ebner, and Stroud. <sup>14</sup> One also has to cite the previous and beautiful analytical work of John and Lubensky <sup>15</sup> who studies the phase transition of disordered granular superconductors near the percolation threshold. They found a more complex H-T phase diagram, consisting of a Meissner followed by an Abrikosov state as the field is increased from zero, whereas a superconducting glass state occurs in the high-field limit induced by frustration effects among loops of the percolation network. However, the authors discussed only the field-independent part of the transition line.

Most of these theoretical predictions are qualitatively reproduced by our experimental results (see Fig. 5), although some discrepancies are worth noticing. First, the models invariably describe direct transitions between the normal state and the superconducting glass state. Experimentally, however, we first observe a transition from normal to a superconducting reversible state, at a critical temperature  $T_c$ , which is practically field independent (see Figs. 1-4, and the phase diagram of Fig. 6). The transition at  $T_c(H)$  occurs clearly below  $T_c(H)$  and separates ergodic and nonergodic superconducting behavior. One may understand the reversible superconducting state as composed by superconducting grains behaving independently from each other. Frustration and irreversibilities appear when links are switched on and closed loops of coupled grains start to form. One should stress, however, that  $T_g(H)$  does not necessarily correspond to the temperature where zero resistance is achieved, as already observed by Carolan et al.9 in magnetoresistance measurements. In a second disagreement with model calculations, we are unable to detect the reversible behavior at low fields generally predicted by the theory. Indeed, one may wonder if a true Meissner state exists in any field range in the oxide superconductors.<sup>26</sup> Finally, comparing the coefficient in Eq. (5) with our data in Table I we obtain values for  $H_0$  ranging from 25 to 100 kG, which are clearly inconsistent with values 1  $kG < 15H_0 < 3$  kG, estimated from the beginning of saturation of  $T_{\rho}(H)$  in Fig. 5. These latter estimated values for  $H_0$  would give an average projected grain area S of about 0.1  $\mu$ m<sup>2</sup>. This size for the mean regions of uniform phase is in fair agreement with prior estimations from experiments on low-field microwave absorption<sup>10</sup> and current-voltage characteristic<sup>27</sup> in ceramic samples. However, this raises the problem of identification of the superconducting grain boundaries, as the calculated grain size is well below the average crystallite size in "123"-type compounds, the latter being typically 1 order of magnitude larger. The possible occurrence of weak links inside crystals, due to twin boundaries or other defects, has been extensively discussed by Deutscher and Muller. 13

It is interesting to compare the experimental values for the parameter a in Table I with the coefficient predicted in the original theory of de Almeida-Thouless. It allows one to estimate the average value for the intergrain coupling J and, from this, to obtain the intergrain current, I=(2e/h)J. Then, a lower bound for the current density across the weak links, j, may be calculated by assuming that the contact area between the grains is given by S. One would finally obtain j>20  $A/\text{cm}^2$ . This is quite reasonable if compared to resistive experimental determinations of the critical current density in Y-Ba-Cu-O ceramic samples in the temperature range close to  $T_c$ .

Another point which may be discussed in relation to spin-glass theory is the striking resemblance of the phase diagrams in Fig. 5 and those presented by magnetic spin-glass systems, <sup>16</sup> mainly concerning the departures from the AT behavior. In the case of spin glasses, the de Almeida-Thouless-type transition has been attributed to a crossover from a high-field Heisenberg regime to a low-field Ising regime induced by local anistropy forces. <sup>16,28</sup> Indeed, for Heisenberg or XY spins, the mean-field theory predicts a spin-glass phase transition occurring along a reversed curvature Gabay-Toulouse line<sup>29</sup>

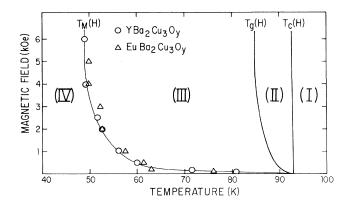


FIG. 6. Schematic high-temperature H-T phase diagram suggested for the high- $T_c$  polycrystalline oxide superconductors. In region I of the phase diagram the system is a normal conductor. In region II the system behaves as a reversible superconductor, probably represented by a collection of disconnected superconducting grains. Region III corresponds to the pure superconducting glass regime and is delimited above by  $T_g(H)$  and below by the  $T_M(H)$  line, which is experimentally determined from the FC susceptibilities of Fig. 4 for the Y-based and Eu-based systems (see text). In region IV the system shows an Abrikosov state, and flux creep effects become increasingly dominant with respect to glassy behavior as the temperature is lowered.

$$H^2 \sim [1 - T_g(H)/T_g(0)]$$
 (6)

One may wonder if a similar crossover occurs in the oxide superconductors, as their description is based on an XY model and so, on the basis of the analogies with spin glasses, one would expect a Gabay-Toulouse-type transition if no anistropy were present. Interestingly, Shih, Ebner, and Stroud<sup>14</sup> in their analysis of the 3D granular model compute the helicity modulus tensor which measures the free-energy increment associated with a twist in the phase at the boundaries of the sample, and found that this quantity is anisotropic with respect to the field direction. In particular, the ratio anistropy/field goes through a maximum in the low-field regime where the AT behavior dominates, decreasing steeply for fields in the saturation  $T_g(H)$  regime. This result indeed suggests an Ising-type transition at low fields. To reinforce this interpretation, a nice  $H^{2/3}$  to  $H^2$  crossover was experimentally found recently in the system Bi-Sr-Ca-Cu-O.<sup>21</sup>

Undoubtedly, one has to be cautious about interpretations of the experimentally observed analogies between oxide superconductors and spin glasses on the basis of the spin-glass theory. The coincidences may be fortuitous, as pointed out by Morgenstern *et al.*<sup>24</sup> Though there are so many that it might be worthwhile to explore this in more detail.

As a final point we would like to comment on the possible relevance and limitations of the "phase glass" state description of superconductivity in the oxide compounds, in comparison with the alternative explanation of irreversible effects in terms of giant flux creep effects in a more conventional Abrikosov state. 19-30 First, we should mention that low-temperature decoration experiments were successful in revealing a triangular vortex lattice in single crystals of Y-Ba-Cu-O.31 This result seems to exclude the granular glass state at least in its simplest version where the penetration length is much larger than the grain's size. Besides, flux trapping experiments may be interpreted in terms of flux creep in the low-temperature range, even for polycrystalline samples.30 On the other hand, at temperatures approaching  $T_c$ , flux decoration fails to reveal a vortex lattice, and there have been reports on flux lattice melting for temperatures neatly below  $T_c$ .<sup>32</sup> Moreover, ac-susceptibility results,<sup>33</sup> experiments on time and temperature dependence of the remanent state,<sup>1,3-5</sup> irreversibilities in magnetoresistance,9 among other experimental evidences, are more suggestive of a glasslike behavior prevailing at high temperatures. Second, we should compare our results with the AT-like behavior reported by Yeshurun and Malozemoff<sup>19</sup> in a single crystal of Y-Ba-Cu-O. In our samples, the  $T_g(H)$  curves saturate and become nearly field independent at quite low values of the applied field, in accordance with predictions of the superconducting glass theory. In contrast, the AT-like line of Ref. 19 extends to fields up to 4 T. Yeshurun and Malozemoff<sup>19</sup> attributed the magnetic irreversibilities in their single crystal to enhanced flux creep effects and interpreted their AT line as a critical current line, whereas we claim that our  $T_g(H)$  is better understood as denoting a broken ergodicity transition.

Having in mind all of these results and ideas, and on the basis of our dc susceptibility data, we propose the low-field-high-temperature H-T phase diagram outlined in Fig. 6 for describing the high- $T_c$  ceramic superconductors. In region I of this diagram the system is a normal conductor. A quasi-field-independent  $T_c(H)$  transition line separates the normal phase from region II, where the system behaves as a reversible superconductor, probably composed by disconnected superconducting grains. Region II is limited in the lower-temperature side by the crossover  $T_{\sigma}(H)$  line, which marks the onset of broken ergodicity effects, characteristic of a superconducting glass state (region III). This state is bound at lower temperatures by the characteristic line represented by  $T_M(H)$ , which denotes the upper limit for the temperature-independent behavior of the Meissner (FC) susceptibility [see Figs. 3(a) and 3(b)]. The data defining  $T_M(H)$  are collected for the Y-based and Eu-based samples. In the Gd-based system, determination of  $T_M(H)$  is difficult because of the paramagnetic contribution to the susceptibility. We suggest that  $T_M(H)$  represents the high-temperature limit for the stability of an Abrikosov lattice. Above this limit, shielding decreases strongly and the system is driven into the pure glassy state. In region IV of the phase diagram of Fig. 6, the effects associated to the pinned Abrikosov vortex state become increasingly important as the temperature is lowered and may even dominate over the superconducting glassy behavior, as suggested by remanence experiments.

## V. CONCLUSION

To summarize, we have discussed dc magnetic susceptibility results for polycrystalline samples of the high-T<sub>c</sub> superconductors YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, EuBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>, and GdBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>. The measurements were performed using the ZFC-FC prescription of spin glasses, and we focus mainly in the high-temperature range. There, the results allow for the determination of a field-temperature phase diagram, which is a general feature of the ceramic oxide superconductors. The ZFC-FC splitting denotes the onset of irreversibility effects in the susceptibility, and defines a characteristic line,  $T_g(H)$  on the H-T plane, which we interpret as resulting from breakdown of ergodicity in the sense of spin-glass phenomenology. This  $T_{\rho}(H)$  line fits to a de Almeida-Thouless-type expression at low fields and becomes field independent for fields in excess of 1-3 kG, depending on the system. This feature is in accordance with predictions of the granular superconducting "phase glass" model14 and looks quite similar to what is observed in more conventional spinglass systems. From our FC susceptibility results we also propose a low-temperature bound for the prevailing superconducting glass state.

As a general conclusion, our magnetic experiments are giving support to a description of the polycrystalline high- $T_c$  superconductors in terms of a superconducting network where frustration and disorder play the major role. This picture is particularly appropriate for the temperature range approaching  $T_c$ .

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