Pairing transition, coherence transition, and the irreversibility line in granular GdBa$_2$Cu$_3$O$_{7−δ}$

J. Roa-Rojas, R. Menegotto Costa, and P. Pureur

Instituto de Física, Universidade Federal do Rio Grande do Sul, P.O. Box 15051, 91501-970 Porto Alegre, RS, Brazil

P. Prieto

Departamento de Física, Universidad del Valle A.A., 25360 Santiago de Cali, Colombia

(Received 14 September 1999)

We report on electrical magnetoconductivity experiments near the superconducting transition of a granular sample of GdBa$_2$Cu$_3$O$_{7−δ}$. The measurements were performed in magnetic fields ranging from 0 to 500 Oe applied parallel to the current orientation. The results show that the transition proceeds in two steps. When the temperature is decreased we first observe the pairing transition, which stabilizes superconductivity within the grains at a temperature practically coincident with the bulk critical temperature $T_c$. Analysis of the fluctuation contributions to the conductivity shows that the universality class for this transition is that of the three-dimensional (3D)-XY model in the ordered case, with dynamic critical exponent $z = 3/2$. Close to the zeroresistance state, the measurements reveal the occurrence of a coherence transition, where the phases of the order parameter in individual grains become long-range ordered. The critical temperature $T_{co}$ for this transition is close to the point where the resistivity vanishes. A strong enlargement of the fluctuation interval preceding the coherence transition is caused by the applied magnetic field. In this region, a 3D-Gaussian regime and an asymptotic critical regime were clearly identified. The critical conductivity behavior for the coherence transition is interpreted within a 3D-XY model where disorder and frustration are relevant. The irreversibility line is determined from magnetoconductivity measurements performed according to the zero-field-cooled (ZFC) and field-cooled data collected on cooling (FCC) recipes. The locus of this line coincides with the upper temperature limit for the fluctuation region above the coherence transition. The irreversibility line is interpreted as an effect of the formation of small clusters with closed loops of Josephson-coupled grains.

I. INTRODUCTION

Since the early studies on high-temperature superconductors (HTSC’s), the pronounced granular character of sintered samples of these systems has been recognized. In particular, their resistive transition shows a two-stage behavior, which is a distinctive characteristic of granular superconductors. At a higher temperature, which is practically coincident with the bulk critical temperature $T_c$, superconductivity stabilizes in homogeneous and mesoscopic regions of the sample (grains). The long-range superconducting state is achieved at a lower temperature $T_{co}$ through a percolation-like process, which controls the activation of weak links between grains. A detailed study of this process, which has been called the “coherence transition,” is given by Rosenblatt et al.

At the coherence critical temperature $T_{co}$, the fluctuating phases of the Ginzburg-Landau order-parameter in each grain couple into a long-range ordered state and a zeroresistance state sets in. The relevant thermodynamics for this problem is obtained from the phase-glass Hamiltonian,

$$H = -\sum_{i,j} J_{ij} \cos(\theta_i - \theta_j - A_{ij})$$

where $J_{ij}$ is the Josephson energy coupling between grains $i$ and $j$, and $\theta_i$ denotes the phase of the order parameter in grain $i$. The gauge factor $A_{ij}$ is given by

$$A_{ij} = \frac{2\pi}{\Phi_0} \int_i^j A \cdot d\ell,$$

where $\Phi_0$ is the flux quantum and the line integral is evaluated between centers of grains $i$ and $j$. The model represented by Eq. (1) belongs to the 3D-XY class with nontrivial (associated to frustration) disorder. Frustration is primarily introduced by a random distribution of the factors $A_{ij}$. The possibility for negative values of the $J_{ij}$ couplings due to, e.g., the occurrence of $\pi$ junctions, may also introduce frustration, much as in the conventional spin-glass models.

Recently, we have presented electrical conductivity and specific-heat experiments, which give significant support to a description of the coherence transition as a genuine critical phenomenon. In this article, we report on detailed magnetoconductivity measurements at low applied fields in a GdBa$_2$Cu$_3$O$_{7−δ}$ (GBCO) granular superconductor. The results show clearly that the transition to the superconducting state in this system is a two-stage process. In the normal phase, when the temperature approaches $T_c$ from above, we observe that conductivity fluctuations scale as predicted by the Gaussian theory. Then, a crossover occurs to a behavior described by the full-dynamic 3D-XY universality class.

Below the pairing transition, and in the region approaching the zero-resistance state, our magnetoconductivity results can be fitted to power laws of a new reduced temperature, $e_0 = [T - T_{co}(H)]/T_{co}(H)$, where $T_{co}(H)$ is almost coincident with the temperature where resistance vanishes. This behavior is an effect of thermal fluctuations that are precursory to the coherence transition. Large Gaussian and critical regimes related to the coherence transition could be discerned.
Measuring the magnetoresistivity according to the ZFC and FCC prescriptions,\textsuperscript{11} we could also determine the irreversibility line. The locus of this line lies clearly above $T_{co}(H)$ and coincides approximately with the upper temperature limit for observing the fluctuation regimes related to the coherence transition.

II. EXPERIMENT

We have prepared a GBCO polycrystalline sample by the standard solid-state technique. Our procedure includes three intermediate grindings before the final sintering at 860 °C. Part of the obtained pellet was cut into a bar with dimensions $8 \times 2 \times 1$ mm for resistivity measurements, part was used for energy-dispersive x-ray analysis (EDAX) and electron microscopy, and part was ground again for powder x-ray diffraction. The EDAX analysis in several points of the sample surface indicated a rather homogeneous composition. Qualitative electron microscopy allowed the visualization of grains with average size of $5 \mu$ and ellipsoid shape, homogeneously distributed on the examined surface. The mass density of the sample was about 80% of the ideal value, and the x-ray spectrum displayed only the reflections expected for the GBCO orthorhombic structure. The obtained lattice parameters, $a = 3.905(8)$ Å, $b = 3.843(4)$ Å, and $c = 11.721(2)$ Å, are within the range of values reported in the literature.\textsuperscript{12}

Resistivity measurements were performed using a low-frequency, low-current ac technique between the zero-resistance state and room temperature. Special care was taken in the temperature interval close to the transition in order to determine accurately the fluctuation contribution to the conductivity. Magnetic fields of 0, 5, 10, 20, 50, 100, 200, and 500 Oe were applied parallel to the current direction. Studies of the fluctuation magnetoconductivity were carried out when the field was applied in accordance to the FCC procedure. For determining the irreversibility line, experiments were performed sequentially according to the ZFC then the FCC recipes.

Temperatures were measured with a Pt sensor corrected for magnetoresistance effects that allows a resolution better than 2 mK. A large number of resistivity vs $T$ points were recorded while the temperature was continuously increased or decreased across the transition in rates not exceeding 3 K/h. Using a numerical method, we could accurately determine the temperature derivative of the resistivity, $d\rho/dT$, in the interval near the transition.

III. RESULTS

Figure 1 shows the resistive transition of our granular GBCO sample in several applied fields. Panel 1(a) presents the resistivity versus temperature plots, whereas panel 1(b) depicts $d\rho/dT$ in the same temperature range. The two-stage nature of the superconducting transition in this systems is apparent from the results in both plots. The applied fields practically do not affect the shape of the curves in the temperature range above the main peak in $d\rho/dT$, whose position is denoted as $T_p$. This temperature, as shown in Fig. 2, is approximately coincident with the pairing critical temperature $T_c$. Below $T_p$, and down to the zero-resistance state, the resistivity curves are dominated by effects related to granularity and depend strongly on the applied field.

The two-stage character of the resistivity transition in the granular HTSC is rendered still more evident by representing the data in terms of the quantity\textsuperscript{13}

\begin{equation}
\chi_\sigma = -\frac{d}{dT}\ln \Delta \sigma,
\end{equation}

where $\Delta \sigma$ is the fluctuation conductivity, given by

\begin{equation}
\Delta \sigma = \sigma - \sigma_R.
\end{equation}

FIG. 1. Resistive transition of our granular GBCO sample in several fields applied parallel to the current, represented as (a) resistivity vs temperature and (b) $d\rho/dT$ vs temperature. The applied fields of 0, 5, 10, 20, 50, 100, 200, and 500 Oe regularly shift downwards the resistivity curves in the regime approaching the zero-resistance temperature.

FIG. 2. Resistive transition for the same data of Fig. 1 represented as $\chi^{-1}_\sigma$ (see text) as a function of temperature. The straight lines are fits to Eq. (6) and are labeled by the respective critical exponents. The irreversibility temperatures for the studied fields are indicated by dots. The irreversibility line is a fit to Eq. (7).
In Eq. (4), $\sigma = 1/\rho$ is the measured conductivity and the subtracted regular term is estimated by extrapolating the high-temperature behavior $\sigma_T = aT + b$, down to the region of the transition. Plots of $\chi^1_{\sigma}$ as a function of $T$ for several applied fields are shown in Fig. 2. There, one clearly observes distinctive behaviors in the temperature interval dominated by fluctuations in the normal phase above $T_p$ (pairing transition), and in the region describing the approach to the zero-resistance state (coherence transition).

The main purpose of presenting the data in terms of $\chi^1_{\sigma}$ is the study of the asymptotic critical fluctuation regimes. Indeed, assuming that $\Delta \sigma$ diverges as a power law when the temperature approaches $T_c$ from above, that is,

$$\Delta \sigma \sim (T - T_c)^{-\lambda},$$

we obtain that

$$\chi^1_{\sigma} = \frac{1}{\lambda} (T - T_c).$$

Thus, as in the Kouvel-Fisher method for studying critical phenomena, the identification of a linear temperature region in $\chi^1_{\sigma}$ allows the simultaneous determination of $T_c$ and the critical exponent $\lambda$. In Fig. 2, straight-line fits to the $\chi^1_{\sigma}$ data are shown.

Above $T_p$, we observe a power-law regime with the exponent $\lambda^{(P)} = 0.35 \pm 0.02$. This value is consistent with expectations from the 3D-XY model with dynamics given by the model $E$ of Hohenberg and Halperin. This model describes the critical dynamics for the superfluid transition in helium, where the order parameter has two components and the density is a conserved quantity. The dynamical universality class of model $E$ is also expected to be valid for the superconducting transition in extreme type-II systems.

Above this genuine critical regime, it is possible to discern intervals dominated by Gaussian fluctuations, which are not addressed in this article.

When the temperature is decreased below the deep minimum where $T_c$ is located, the $\chi^1_{\sigma}$ data of Fig. 2 show the characteristic behavior of granular samples. First, $\chi^1_{\sigma}$ increases steeply, such that its shape is nearly symmetrical with respect to the high-temperature side of the pairing transition, until a field-dependent maximum is attained. Then, $\chi^1_{\sigma}$ decreases to zero at $T_{co}(H)$ with a curvature that is inverted when compared to that observed above $T_c$. This latter behavior results from the effect of phase fluctuations in the granular array above the coherence transition, and characterizes the so-called paracoherent state. When the field is increased, the width of the paracoherent fluctuation interval is correspondingly enlarged.

Two power-law regimes with field-independent exponents are identified in the paracoherent region of $\chi^1_{\sigma}$. Farther from $T_{co}$, and just below the granularity-induced maximum in $\chi^1_{\sigma}$, we obtain the exponent $\lambda^{(c)} = 0.53 \pm 0.03$. The identification of this regime becomes clearer when the field is increased above 20 Oe. Also evident is the asymptotic regime near $T_{co}(H)$, which is characterized by the exponent $\lambda^{(c)} = 4.2 \pm 0.2$.

Figure 3(a) shows an example of magnetoresistance measurements performed in the ZFC and FCC conditions. The two curves split below a characteristic temperature $T_{irr}(H)$, which marks the onset of irreversible effects in the magnetotransport properties. Figure 3(b) shows the difference between the FCC and ZFC magnetoresistances plotted as functions of temperature. Such plots allow an accurate determination of $T_{irr}(H)$. In the range of studied fields, the irreversibility temperatures so determined are coincident with those obtained from ZFC and FCC dc-magnetization measurements. The irreversibility temperatures vary with the applied field according to

$$\left( \frac{H}{H_0} \right) = \left[ 1 - \frac{T_{irr}(H)}{T_{irr}(0)} \right]^\gamma,$$

where $\gamma = 1.48 \pm 0.05$ and $H_0 = 63 \pm 2$ kOe.

The experimentally determined $T_{irr}(H)$, which are signaled by full circles in Fig. 2, are nearly coincident with the temperatures of the granular maximum in $\chi^1_{\sigma}$. This behavior clearly associates the onset of irreversibility effects in our granular superconductor with a threshold that marks the upper temperature limit of the paracoherent interval and separates single grain from collective grain responses in the fluctuation conductivity.

Rather interesting is the behavior of $\chi^1_{\sigma}$ just above $T_{irr}(H)$ and up to the deep minimum where the pairing temperature is located. In this temperature range $\chi^1_{\sigma}$ is approximately symmetrical with respect to the behavior observed in the normal phase just above $T_c$. Indeed, as shown in Fig. 2, in a certain region of this interval $\chi^1_{\sigma}$ may be fitted to a straight line as $\chi^1_{\sigma} = (1/\lambda^-)(T_c - T)$, where $T_c$ is nearly coincident with $T_c$ and $\lambda^- = 0.3$. This suggests an unexpected symmetry of the fluctuation-conductivity divergence around $T_c$. We note, however, that neither the $\lambda^-$ regime nor the coherence transition have been identified in single
crystal samples, so that the \( \lambda^- \) regime might be related to finite-size effects in isolated grains of the ceramic HTSC samples.

IV. DISCUSSION

A. Pairing transition

The critical exponent for fluctuation conductivity is given by

\[
\lambda = \nu \left( 2 + z - d + \eta \right),
\]

where \( \nu \) is the critical exponent for the coherence length, \( z \) is the dynamical critical exponent, \( d \) is the dimensionality, and \( \eta \) is the exponent of the order-parameter correlation function.

When the temperature approaches close enough to \( T_c \), the critical thermodynamics of the superconducting transition is well described by the 3D-XY universality class. According to renormalization-group calculations, in this case one would expect \( \nu = 0.67 \) and \( \eta = 0.3 \). On the other hand, the theory of dynamical critical scaling predicts \( z = 3/2 \) for the superconducting transition. Substituting these values in Eq. (8), one obtains \( \lambda \cong 0.35 \), in agreement with our experimental finding for the asymptotic fluctuation regime prior to the pairing transition, as shown in Fig. 2. This regime is observed in the whole range of applied fields and extends up to 0.85 K above \( T_c \).

B. Coherence transition

The power law behavior of the conductivity, characterized by the exponent \( \lambda_{cr}^{(c)} = 4.2 \), which governs the approach to the zero-resistance state at \( T_{co} \), is interpreted as an effect of genuine critical fluctuations precursory to the coherence transition. In contrast to the critical regime near the pairing transition, the asymptotic critical interval in the paracoherent transition is large enough, the asymptotic conductivity regime generally corresponds to \( \lambda_{cr}^{(c)} \cong 4 \). This indicates that the dynamical universality class of the coherence transition in the presence of a small magnetic field is closer to that of a spin glass, where one indeed expects \( z \cong 4 \).

Farther from \( T_{co} \), and just below the maximum in \( \chi_{cr}^{-1} \), the regime described by the exponent \( \lambda_{cr}^{(c)} \cong 0.5 \) is likely due to three-dimensional Gaussian fluctuations. Using the mean-field values \( \nu = 0.5 \), \( \eta = 0 \), and \( z = 2 \) in Eq. (8), one reproduces the measured value of \( \lambda_{cr}^{(c)} \). We notice that this is probably the first identification of a Gaussian regime corresponding to a vortex-glass-like transition. Such fluctuations were searched for unsuccessfully in I-V experiments in YBCO thin films. It is also interesting to note that the fluctuation spectrum is isotropic in the whole paracoherent interval, in spite of the strong planar anisotropy of the intra-grain superconductivity.

The shape of the \( \chi_{cr}^{-1} \) results of Fig. 2 in the paracoherent region strongly suggests that the whole measurements may be scaled into a single curve.

The scaling behavior proposed for the fluctuation conductivity close to a vortex-glass transition is given by

\[
\Delta \sim H^{-1/2 \left( 2 + z - d \right)} S_{\pm} \left( \frac{\varepsilon_0}{H^{1/2}} \right),
\]

where \( \varepsilon_0 = (T-T_{co})/T_{co} \), and \( S_{\pm} \) are scaling functions above and below \( T_{co} \), respectively.

Assuming, following Fisher et al., that

\[
[T_c - T_{co}(H)] \sim H^{1/2 \epsilon}
\]

and using the scaling variable introduced by Kötzler et al., we derive that

\[
\frac{d}{d\tau} \ln S_{\pm} (\tau) = \chi_{cr}[T_c - T_{co}(H)]
\]

in the fluctuation region above the coherence transition and below the pairing transition. Figure 4 shows a plot of \( \chi_{cr}[T_c - T_{co}(H)] \) vs \( \tau \) determined from the results presented.
in Fig. 2. Good scaling is indeed obtained in most of the range [0,1] for the variable τ. Deviations occur close to the high-temperature end because of the steep variation of χσ⁻¹.

C. Irreversibility line

The de Almeida-Thouless-like behavior of the irreversibility line in our granular HTSC at low applied fields may be explained by the phase-glass model of Eq. (1). The coincidence of the irreversibility temperature at a given field with the maximum in χσ⁻¹, signaling the high-temperature limit of the paracoherent fluctuation interval, is a clear indication that the onset of irreversibility effects is a threshold separating single-grain from collective-grain behavior. The Hamiltonian of Eq. (1) predicts that when loops of weakly coupled grains are formed, frustration may occur since the coupling energies between all pairs of grains cannot be simultaneously minimized, in the presence of the random gauge factors Aij. As a consequence, the state of a small aggregate of interacting grains becomes highly degenerate with many nearly equivalent low-energy configurations. Irreversible effects occur when the system evolves through the rugged landscape characteristic of the phase space in disordered and frustrated systems. Thus, the irreversibility line at low fields in granular superconductors is a consequence of the stabilization of relatively small aggregates of coupled grains, and do not represent necessarily a phase transition. In other terms, the irreversibility line is related to the onset of short-range ordering, in contrast to the percolation-like coherence transition that is achieved when an (ideally) infinite cluster of coupled grains is formed in the sample.

V. CONCLUSIONS

Our low-field magnetoconductivity results in a granular GBCO superconductor show that the resistive transition in this system is a two-stage process. When the superconducting state is approached from above we first observe the occurrence of a pairing transition, which stabilizes the condensate within the grains at a critical temperature TC, close to that of a bulk specimen. This transition is preceded by large regimes dominated by fluctuations of the order parameter amplitude. Immediately above TC, a full dynamic 3D-XY fluctuation regime, with z = 3/2, is clearly identified. This regime extends up to 0.85 K above TC, and is robust against magnetic fields applied parallel to the current in the range 0–500 Oe.

The genuine superconducting state with zero-resistance stabilizes at the coherence transition that occurs at a lower temperature Tco, where a long-range-ordered state for the phase of the order parameter is established in the whole granular array. The approach to the zero-resistance state is dominated by strong fluctuations of the phase of the order parameter of individual grains. Extended power law regimes corresponding to 3D-Gaussian and critical fluctuations are identified in the magnetoconductivity experiments. The obtained exponent in the asymptotic critical regime indicates that the static and dynamic universality classes for the coherence transition is that of the 3D-XY model described by the phase-glass Hamiltonian of Eq. (1), where the disorder is nontrivial (accompanied by frustration) and critically relevant. A scaling analysis of the conductivity in the interval between Tco and TC shows that the coherence transition in our homogeneously granular superconductor fits into the more general framework of the vortex-glass theory.

Magneetoconductivity measurements performed according to the ZFC and FCC prescriptions allowed the determination of the irreversibility line. The locus of this line lies clearly above the zero-resistance temperatures and coincides approximately with the high-temperature limit for the fluctuation region above the coherence transition. This indicates that the low-field irreversibility line in granular superconductors is a threshold separating single-grain from collective-grain behavior and is related to the formation of small clusters with closed loops of Josephson-coupled grains.

ACKNOWLEDGMENTS

We are indebted to Dr. Miguel A. Gusmão for a careful reading of the manuscript and to Dr. Enzo Granato for illuminating discussions. We also acknowledge Dr. M. Z. Vascconcellos for the EDAX analysis and Claudia M. Haetinger for help with the text. This work was partially supported by the Brazilian Ministry of Science and Technology under Contract PRONEX/FINEP/CNPq No. 41.96.0907.00. One of us (J.R.R.) received support by CNPq (Brazil) and COLCIENCIAS (Colombia).

9See, for example, K. Binder and A. P. Young, Rev. Mod. Phys. 58, 801 (1986).