



AC susceptibility study of the intergranular irreversibility line in BSCCO ceramic superconductors

J.L. González^{*}, P. Muné, L.E. Flores, E. Altshuler

Superconductivity Laboratory, IMRE, University of Havana, 10 400 La Habana, Cuba

Received 18 August 1995

Abstract

The intergranular irreversibility line of granular BSCCO-2223 sample was found by measuring the AC susceptibility. The data are compared with the analogous ones for a YBCO ceramic. The measured values agree with the ones determined through the giant flux-creep theory adapted to the intergranular case, choosing appropriate parameters. This result suggests that the change from non-dissipative to dissipative states can be regarded as an intergranular pinning–depinning transition.

1. Introduction

In the H – T phase diagram of a high- T_c superconductor (HTSC) the existence of a feature called “irreversibility line” is well established. Below this line, a finite critical current density exists, while it turns to zero above, corresponding to irreversible and reversible magnetic behaviors, respectively. This was initially reported by Müller et al. [1] in 1987 and it has been the subject of many scientific efforts up to now. Various methods have been used to determine this line such as the DC magnetization [1], AC susceptibility [2], magnetization–magnetic field loops [3], and so on. The origin of this boundary has been attributed to depinning (giant flux creep [4]), the melting of the vortex lattice into a vortex fluid due to thermal fluctuations (flux-lattice melting [5]) and the transition from the vortex-liquid into vortex-glass [6].

In the case of ceramic superconductors, the phase diagram is more complex, because the system can be

regarded as an array of superconducting grains interconnected by weak links [7]. Consequently, one can talk about intragranular or intergranular properties, making it possible to define an intergranular irreversibility line above which the transport critical current disappears [8].

In this work we found the intergranular irreversibility line of a Pb doped Bi-2223 ceramic through complex susceptibility measurements and we compared the results with the model of giant flux-creep of Malozemoff adapted to the intergranular case.

2. Theory

When a magnetic field is applied to the sample it penetrates firstly the intergranular regions, producing Josephson vortices. On the other hand, it has been proved that the principal dissipation process occurs due to the movement of these vortices [9]. If a granular material is regarded as an array of cubic

^{*} Corresponding author.

grains interconnected by weak links, it can be shown that the depth of the pinning centers located at the corners between four grains is proportional to the intergrain coupling energy, E_j [10]. As a consequence of this, Müller [11] developed a model where the intergranular pinning energy is proportional to the average grain boundary Josephson junction current, $I_0(T, H)$, and has the form

$$U_0 = \frac{\phi_0}{2\pi} \beta I_0(T) \frac{H_0(T, r_g)}{H_0(T, r_g) + H_i} \quad (1)$$

Here, ϕ_0 is the flux quantum, and $H_0 = (\phi_0/4\mu_0\lambda_g r_g)$, where r_g is the grain radius, β is a factor that accounts for inhomogeneities of the junctions, λ_g is the London penetration depth of the grains, and H_i is the effective intergranular field felt by the junctions. Many papers have been devoted to the study of H_i , which can be expressed as [12]

$$H_i = H_{ap} - GM(H_i), \quad (2)$$

where H_{ap} is the applied field, $M(H)$ is the field-dependent magnetization of the grains, and G is a geometric factor which depends on the sample's microstructure. It should be remarked that, as pointed out by Ref. [13], the statistical distribution of G in typical Bi–Pb–Sr–Ca–Cu–O ceramics comprises both positive and negative values of G , with a peak at some positive value of G close to zero. Thus, $H_i \approx H_{ap}$.

The giant flux-creep theory focuses the role of thermal activation in the determination of the critical current density through the relation

$$J_c = J_{c0} \left[1 - \frac{kT}{U_0} \ln \left(\frac{Bd\Omega}{E_c} \right) \right], \quad (3)$$

where J_{c0} is the critical current density in the absence of thermal activation, B is the magnetic induction, d is the distance between pinning centers, E_c is a minimum measured electric field, Ω is a typical oscillation frequency and k is the Boltzman constant. The thermal activation causes J_c to drop below the measurement threshold and the condition $J_c = 0$ for this criterion gives the irreversibility line. In agreement with the latter, the intergranular irreversibility line can be obtained if the parameters in Eq. (2) are substituted by the intergranular ones. By

substituting Eq. (1) in Eq. (3), and making $J_c = 0$, $d = r_g$ and $H_i = H_{ap}$ we obtain

$$\frac{\phi_0 \beta I_0(T)}{2\pi} \frac{H_0(T, r_g)}{H_0(T, r_g) + H} = kT \ln \left(\frac{H \langle r_g \rangle \Omega 10^{-4}}{E_c} \right), \quad (4)$$

where H and H_0 are given in Oe, r_g in m, Ω in Hz, E_c in V/m, $I_0(T)$ in A and T in K.

It should be noted that, in the condition $H_i = H_{ap}$, we implicitly assume that the geometric factor G is negligible. The theoretical dependence $H_{irr}(T)$ was obtained solving Eq. (4) by means of computational methods.

3. Experimental

The sample used in this work was obtained by means of a common solid-state reaction described elsewhere [14]. The starting powders were Bi_2O_3 , PbO , SrCO_3 , CaCO_3 and CuO mixed in appropriate proportions producing the final compound $\text{Bi}_{1.6}\text{Pb}_{0.4}\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_8$. The pellets were sintered at 850°C during 40 h and cut as slabs of typical dimensions $2.6 \times 0.7 \times 10 \text{ mm}^3$. The critical current density was measured through a standard four-probe technique, obtaining at 80 K in zero magnetic field the value of 130 A/cm^2 .

The AC susceptibility was recorded using a standard susceptometer, i.e. a drive coil with two compensated pickup coils, and the sample situated into one of them. The signal obtained (which was proportional to the susceptibility) was processed with a phase-sensitive detector, sampling the real and imaginary parts. All the measurements were performed at a frequency of 1 kHz and with a AC amplitude of about 0.1 mT. The temperature was controlled with a Lake Shore 330 autotuning temperature controller and the DC magnetic field was moved in the interval from 0 to 200 Oe.

4. Results and discussion

Figs. 1(a) and (b) show the temperature dependence of the resistivity and transport critical current

of the sample at zero applied field. The resistive transition was broad, which can be interpreted in terms of the existence of a distribution of “qualities” of the intergrain Josephson junctions. Its onset was at

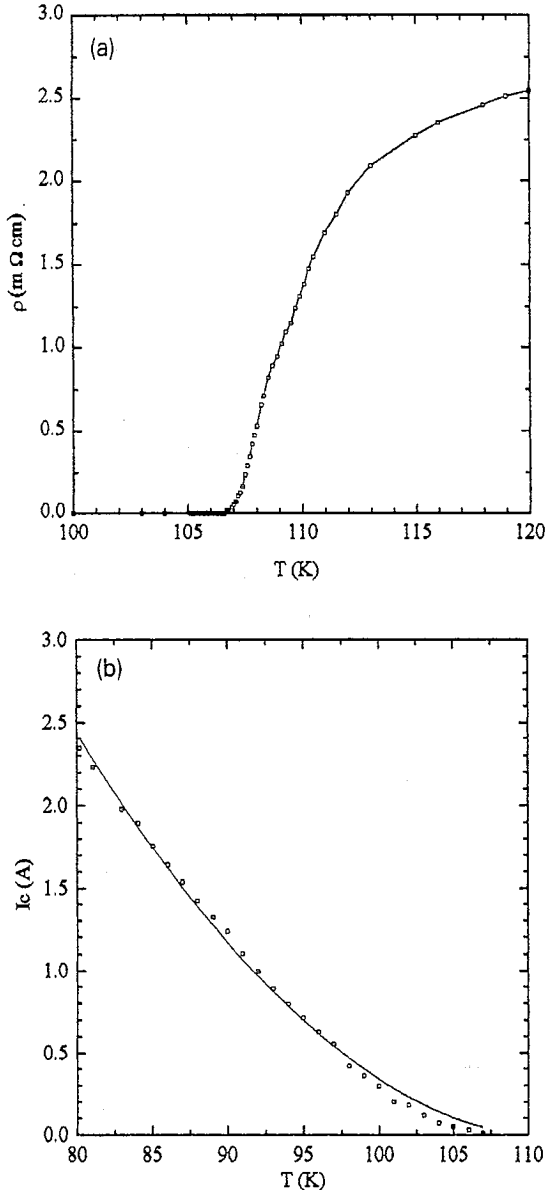


Fig. 1. (a) Resistivity vs. temperature curve of the BSCCO polycrystal. (b) Temperature dependence of the transport critical current for the BSCCO polycrystal. The solid line is a fit to the expression $I(T) = I_0(1 - T/T_{c_j}[0])^n$, with $I_0 = 26$ A, $T_{c_j} = 110.6$ K and $n = 1.9$.

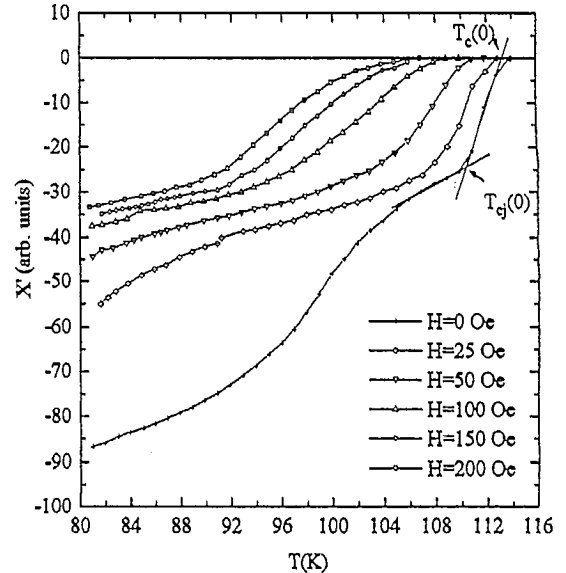


Fig. 2. In-phase component of the magnetic AC susceptibility of the BSCCO polycrystal for different DC applied fields.

about 114 K. Fig. 1(b) also shows a theoretical fit to the experimental data according to $I_{c_i}(T) = I_0(0)(1 - T/T_{c_j}[0])^n$ where $I_0(0) = 26$ A and $n = 1.9$ were used as fitting parameters, while $T_{c_j}[0]$ is a zero-field Josephson critical temperature to which we refer later [15]. The value of n indicates that the intergrain junctions of our sample behave as SNS ones, as previously reported in the literature [15].

The temperature dependence of a typical data set of the real part of the complex susceptibility for different DC magnetic fields can be viewed in Fig. 2. The curves present two transitions which were attributed to the inter- and intragranular cases [16]. In our measurements, for zero applied field, one can distinguish the one associated to the junctions at approximately 110.6 K, T_{c_j} , while the transition associated to the grains occurs at approximately 113.3 K in good agreement with the one found by means of the resistivity measurement. $T_{c_j}(0)$ was taken at the intercept of the linear fits of χ' at $H_{ap} = 0$ in the intervals 100–110 K and 111–113 K, respectively. $T_c(0)$ was taken as the intercept of the last fit with the $\chi' = 0$ axis, as depicted in Fig. 2.

At the same time we could find two peaks in the case of the imaginary part of the complex susceptibility; the ones found at lower temperatures are

displayed in Fig. 3 which shows the temperature dependence of the out-of-phase signal for different DC fields. The temperature at which this peak occurs, T_d , was regarded as the irreversibility temperature. Although this last assessment has been disputed in the literature [17] it seems to be that, in the case of small AC field superimposed upon a large DC field, the T_d temperature coincides with the irreversibility temperature found by other methods, e.g. DC magnetization [2].

At this point it is straightforward to compare the experimental results with the model suggested in the theory. This is illustrated in Fig. 4, where a 'ditional experimental data corresponding to a YBCO sample taken from Ref. [8] are also plotted.

In order to fit our BSCCO data with the model we solved Eq. (4) through computational methods. For the temperature dependence of I_0 we used $I_0(T) = I_0(0)[1 - T/T_c(0)]^2$, typical for a SNS junction, and in the case of the penetration depth [19], $\lambda(T) = \lambda(0)[1 - (T/T_c(0))^4]^{-0.5}$ with $\lambda(0) = 0.2 \mu\text{m}$.

We selected $\beta\phi_0 I_0(0)/2\pi = 7.8 \text{ eV}$, $E_c = 10^{-3} \text{ V/m}$, and, for r_g , we used a triangular distribution with $r_{g \text{ min}} = 0.5 \mu\text{m}$, $r_{g \text{ peak}} = 2 \mu\text{m}$ and $r_{g \text{ max}} =$

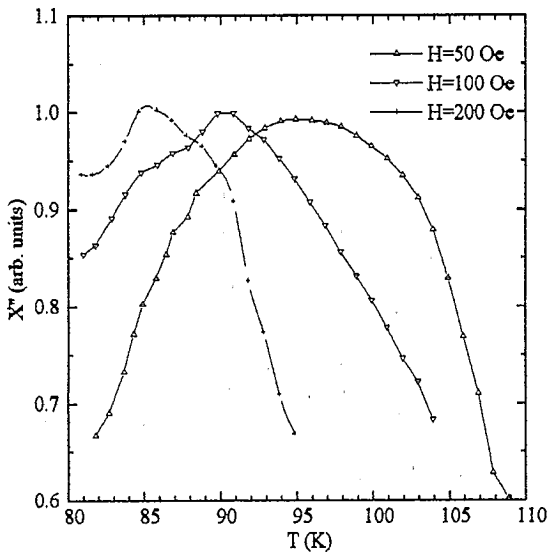


Fig. 3. Normalized out-of-phase component of the magnetic susceptibility of the BSCCO polycrystal for three different DC applied fields.

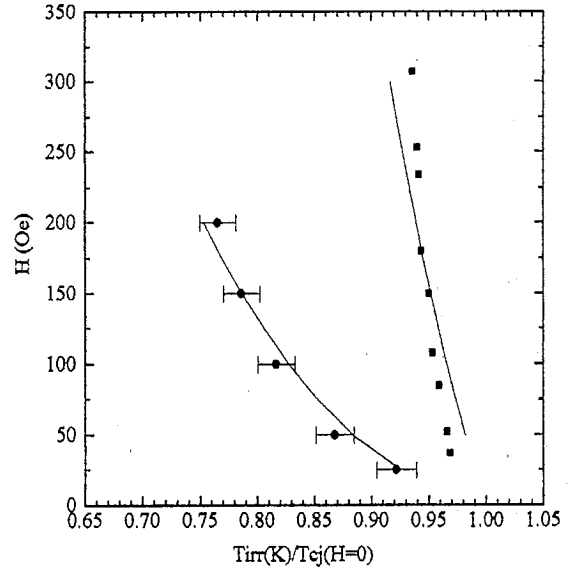


Fig. 4. Irreversibility fields vs. reduced temperature for a BSCCO polycrystal (solid circles) and for a YBCO polycrystal (solid squares) sample. The solid line represents fits according to the model proposed.

$5 \mu\text{m}$. We regarded (r_g) as the average value of r_g of our distribution, i.e., $2.5 \mu\text{m}$. In the case of vortices oscillating in intragranular pinning centers, like oxygen vacancies, Ω has been taken between 10^8 and 10^{12} Hz [20]. Here, as we are talking about intergranular pinning centers which are bigger than the intergranular ones, a value 10^6 Hz was assigned to Ω .

In the case of YBCO we chose the same temperature dependence of $\lambda(T)$ but, with $\lambda(0) = 0.15 \mu\text{m}$, $\beta\phi_0 I_0(0)/2\pi$ in this case was 24.7 eV and, since the experimental data were obtained in FC conditions, H_i was taken as the applied field [19]. For r_g we used the same triangular distribution for the BSCCO ceramic, $E_c = 10^5 \text{ V/m}$, $\Omega = 10^6 \text{ Hz}$, $T_c(0) = 92 \text{ K}$, $T_{c_j}(0) = 91.4 \text{ K}$ and, for the temperature dependence of I_0 , we used a relation of the type $I_0(T) = I_0(0)[1 - T/T_c(0)]^1$ [21].

Two features can be observed in Fig. 4. The first is that, as expected, the irreversibility field decreases when the temperature increases and, on the other hand, the BSCCO line is less tilted than the one for the YBCO compound. This last behavior has been

reported in the case of intragranular measurements in these samples [22].

One also observes in Fig. 4 a good overlapping between the data and the solid lines that represent the solutions of Eq. (3) for the parameters mentioned earlier. This implies that the change from zero resistance to a dissipation state with a critical current can be understood as a pinning–depinning transition of Josephson vortices. The pinning of these vortices occurs due to inhomogeneities in the coupling energy between the grains [23].

Different parameters were tried in order to obtain a good matching between theory and experiment. In particular, we noted that good fits were possible only if the BSCCO junctions were regarded as SNS, which implies that the exponent in the temperature-dependent critical current, $I_0(T)$, should be taken as $n = 2$. The same occurred for YBCO, but with $n = 1$, typical for SIS junctions. Then, it seems that the shape of the line in the H – T plane, according to the model, strongly depends on the junction type.

Although the solutions of Eq. (4) reproduced the experimental data, we think that, if other details are considered, even better results can be achieved. Among these, the most important might be the fact that the junctions do not have all the same values of $I_0(0)$, which means that a distribution function for this parameter should be considered.

5. Conclusions

The intergranular irreversibility line for a BSCCO-2223 system was measured through AC susceptibility with a superimposed DC field. A comparison between BSCCO and YBCO experimental data resembles the case of intragranular measurements for the same two compounds. This behavior can be explained using the giant flux-creep theory conveniently adapted to the intergranular case with an adequate choosing of parameters. The shape of the irreversibility line in the H – T plane strongly depends on the type of junctions involved.

References

- [1] K.A. Müller, M. Takashige and J.G. Bednorz, *Phys. Rev. Lett.* 58 (1987) 1143.
- [2] R.B. Flippen, *Phys. Rev. B* 45 (1992) 12 498.
- [3] U. Welp, G.W. Crabtree, J.L. Wagner, D.G. Hinks, P.G. Radaelli, J.D. Jorgensen and J.F. Mitchell, *Physica C*, in press.
- [4] Y. Yeshurun and A.P. Malozemoff, *Phys. Rev. Lett.* 60 (1988) 2202.
- [5] D.R. Nelson, *Phys. Rev. Lett.* 60 (1988) 1973;
D.R. Nelson and H.S. Seung, *Phys. Rev. B* 39 (1989) 9153.
- [6] M.P.A. Fisher, *Phys. Rev. Lett.* 62 (1989) 1415;
D.S. Fisher, M.P.A. Fisher and D.A. Huse, *Phys. Rev. B* 43 (1991) 130.
- [7] D.H. Kim, K.E. Gray, R.T. Kampwirth and D.M. McKay, *Phys. Rev. B* 42 (1990) 6249.
- [8] L. Miu, *J. Supercond.* to be published.
- [9] L. Miu, S. Popa, A. Crisan, G. Aldica and J. Jacklowski, *J. Supercond.* 6 (1993) 279.
- [10] C.J. Lobb, D.W. Abraham and M. Tinkham, *Phys. Rev. B* 27 (1983) 150.
- [11] K.H. Müller and M. Nikolo, *Phys. Rev. B* 43 (1991) 7976;
K.H. Müller, *Physica C* 168 (1990) 585.
- [12] E. Altshuler, J. Musa, J. Barroso, A.R.R. Papa and V. Venegas, *Cryogenics* 33 (1993) 308.
- [13] P. Muné, E. Altshuler, J. Musa, S. Garcia and R. Riera, *Physica C* 226 (1994) 12.
- [14] K. Jain, D.K. Suri, K.C. Nagpal, S.U.M. Rao and B.K. Das, *Jpn J. Appl. Phys.* 29 (1990) 576.
- [15] L.A. Angurel, F. Lera, C. Rillo and R. Navarro, *Physica C* 230 (1994) 361.
- [16] R.B. Goldfarb, M. Leleth and C.A. Thompson, *Magnetic susceptibility of superconductors and other spin systems*, (Plenum, New York, 1992).
- [17] S. Samarappuli, A. Schilling, M.A. Chernikov, H.R. Ott and Th. Wolf, *Physica C* 201 (1992) 159.
- [18] D.R. Harshman, G. Aeppli, E.J. Ansaldo, B. Batlogg, J.H. Brewer, J.F. Carolan, R.J. Cava, M. Cellio, A.C.D. Chaklader, W.N. Hardy, S.R. Kreitzman, G.M. Luke, D.R. Noakes and M. Senba, *Phys. Rev. B* 36 (1987) 2386.
- [19] Y. Yang, C. Beduz, Z. Yi and R.G. Scurlock, *Physica C* 201 (1992) 325.
- [20] C.W. Hagen and R. Griessen, in: *Studies of High-Temperature Superconductors (vol III)* (Nova Science, Commack 1989).
- [21] N. Savvides, *Physica C* 165 (1990) 371.
- [22] A. Umezawa, W. Zhang, A. Gurevich, Y. Feng, E.E. Hellstrom and D.C. Larbalestier, *Nature (London)* 364 (1993) 129.
- [23] J.R. Clem, *Physica C* 153–155 (1988) 50.