Vol. **109** (2006) *ACTA PHYSICA POLONICA A* No. 4–5

Proceedings of the XI National School "Collective Phenomena and Their Competition"
Kazimierz Dolny, September 25–29, 2005

Superconducting Transition in Granular Bi–Sr–Ca–Cu–O Superconductors

M. Gazda[a], B. Kusz[a], W. Sadowski[a], S. Stizza[b], R. Natali[b] and V. Di Stasio[b]

[a]Faculty of Applied Physics and Mathematics
Gdańsk University of Technology
Narutowicza 11/12, 80-952 Gdańsk, Poland
[b]Dipartimento di Matematica e Fisica, Università di Camerino, INFM, Italy

In this work we study the superconducting transition in the samples containing relatively small granules of high-T_c Bi–Sr–Ca–Cu–O superconductors. The samples were obtained by the glass-ceramic technology. Two-stage character of the superconducting transition was studied. Pairing transition temperature depends on the magnetic field. Coherence transition is very sensitive to the current flowing through the sample, magnetic field and the phase composition and microstructure of the sample. The analysis of superconducting transition by means of the logarithmic derivative of the fluctuation conductivity shows the mesoscopic granularity of the samples.

PACS numbers: 74.62.–c, 74.81.Bd, 74.72.Hs

1. Introduction

Solid state crystallisation of the $(Bi_{0.8}Pb_{0.2})_4Sr_3Ca_3Cu_4O_x$ glass leads to the formation of a granular material composed of oxide superconductors belonging to the bismuth family embedded in the non-superconducting matrix forms [1].

Typical examples of a superconducting transition in disordered, granular material containing a small amount of relatively small superconducting granules are shown in Fig. 1. A granular material transits into the superconducting state in a two-step process [2]. First, at temperature T_1 so-called pairing transition occurs, that is, most of the isolated granules transit into the superconducting state. The second step, the coherence transition, appears at temperature T_2 when the grains couple into a long-range ordered state. In this work we study the superconducting transition in the samples containing mainly small granules of the $Bi_2Sr_2CaCu_2O_x$ (2212) superconductor.

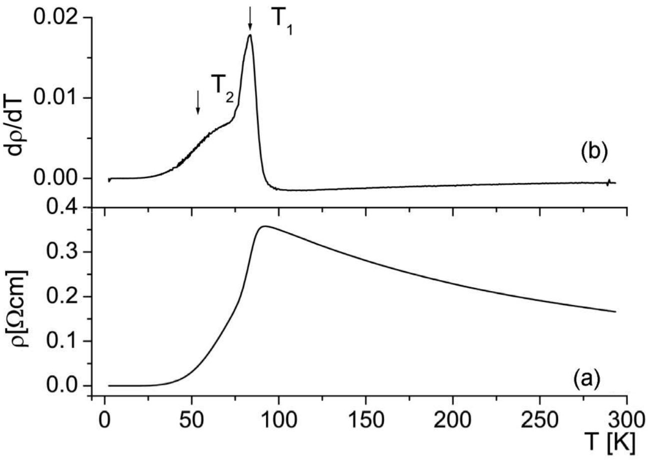

Fig. 1. An example of the superconducting transition in granular material (a), as well as an illustration of determination of two critical temperatures (b) (T_1 — pairing transition temperature, T_2 — coherence transition temperature).

2. Experimental

In order to obtain a granular material short times of annealing were applied to the $(Bi_{0.8}Pb_{0.2})_4Sr_3Ca_3Cu_4O_x$ glass (830°C, 1 min; and 860°C, 2 min). The phase composition of the samples was studied by X-ray diffraction. The dimensions of the 2212 crystallites were estimated by X-ray diffraction and scanning electron microscopy (SEM). The average radii of the granules are 50 and 300 nm in the sample annealed at 830°C and 860°C, respectively. The details of the experimental procedures are published elsewhere [3].

The measurements of temperature dependence of resistivity were carried out in the temperature range from 3 K to 300 K. The measurements were performed with current densities between 0.3×10^{-4} A/cm^2 and 0.3 A/cm^2 (current between 1 μA to 10 mA). In the case of the sample annealed at 860°C for 2 min weak magnetic field (up to 2 T) was applied.

3. Results and discussion

Figure 2 shows the temperature dependence of resistivity (Fig. 2a) and its derivative (Fig. 2b) of the sample annealed at 830°C for 1 min. The sample contains only a few percent of the 2212 phase dispersed in oval granules not larger than 50 nm. Nevertheless, the transition to the superconducting phase is quite sharp. The pairing transition connected with the presence of the granules of 2212 superconductor is represented by a maximum seen in Fig. 2b between 84.0 and 85.5 K. The maximum is much smaller and wider than in typical ceramic superconductors. This reflects the fact that the sample contains only a small amount of the 2212 phase, dispersed in small granules. The material inside the granules, because of a short annealing time, is defected, hence there is a distribution of critical temperatures which widens the pairing transition. Its position (around 85 K) does not change with the current. The value of transition temperature is typical of the 2212 phase, which means that the size of the granules is still too large to influence the critical temperature. Size-dependent critical temperature was observed in various superconductors when the size of grains was of the order of 5–50 nm (e.g. Bi and MgB_2) [4].

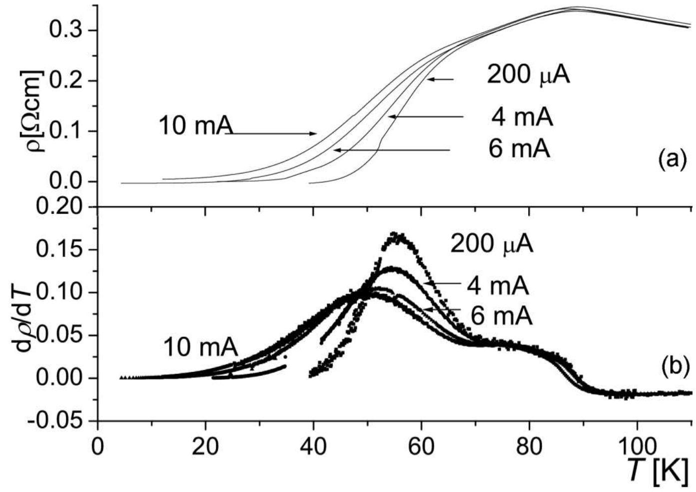

Fig. 2. Temperature dependence of resistivity of the sample annealed at 830°C for 1 min measured with the current between 200 μA and 10 mA (a) and temperature dependence of the derivative $d\rho/dT$ (b).

The coherence transition temperature depends strongly on the current flowing through the sample. Intergranular critical current is relatively small, so that even the current as small as 1 mA causes a visible shift of the coherence transition towards the lower temperatures. This temperature is determined by the links between the grains, current density, and magnetic field [5].

Figure 3 shows the superconducting transition in a glass-ceramic sample containing the granules of about 0.3 μm. The measurements were carried out at small magnetic field applied in the direction perpendicular to the sample surface. The values of transition temperature are collected in Table. It can be seen that the pairing transition temperature shifts towards lower temperatures with the magnetic field increase. Magnetic field penetration depth in the 2212 superconductors is about 200 nm [6], which is of the same range that the average radius of superconducting granules in our samples. Therefore, a large number of the granules are very sensitive even to a relatively small magnetic field. The shape of the superconducting transition at magnetic field larger than 0.125 T is qualitatively different from that observed without the field.

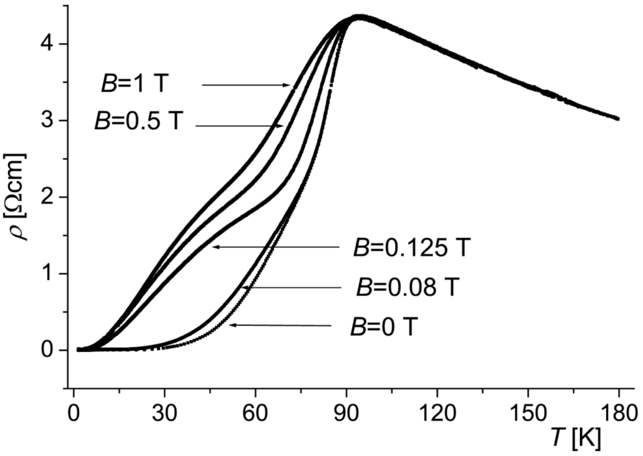

Fig. 3. The superconducting transition in a glass-ceramic sample measured at magnetic field applied in the direction perpendicular to the sample surface ($B \leq 1$ T).

TABLE

Transition temperatures of the sample annealed at 860°C for 2 min as a function of the magnetic field.

B [T]	Onset temperature	Pairing transition temperature	Coherence transition temperature	s
0	92.6	85	60.8	4.3 ± 0.2
0.08	92.5	84.4	60.2	4.2 ± 0.2
0.125	92.5	81.7	36.7	1.9 ± 0.3
0.5	92.5	76.6	22.5	1.8 ± 0.3
1	91.7	72.8	21.5	2.0 ± 0.3
2	92	70.6	21.2	1.8 ± 0.3

The coherence transition temperature decreases very rapidly with the increase in magnetic field. Such behaviour is typical of the system containing a large amount of weak links between the grains.

The two-stage character of the transition to the superconducting state in granular superconductors is even more evident if the data are plotted in terms of the quantity

$$X = -\frac{\mathrm{d}(\ln \Delta\sigma)}{\mathrm{d}T}, \tag{3.1}$$

where $\Delta\sigma$ is the fluctuation conductivity $\Delta\sigma = \sigma - \sigma_R$. σ is the measured conductivity whereas σ_R is the conductivity in normal state, estimated on the basis of high-temperature behaviour [2]. Assuming that the superconducting transition is of second order, the development of the long-range correlations may be expressed as a power divergence of the coherence length in the vicinity of the transition. Taking $\Delta\sigma$ as a measure of the long-range correlations, we may express it as: $\Delta\sigma \propto \xi^{d-z-2}$, where d is the dimensionality of the system and z is the dynamical exponent. Knowing that

$$\xi \propto \left(\frac{T - T_{\mathrm{c}}}{T_{\mathrm{c}}}\right)^{\nu}, \tag{3.2}$$

where ν is the static exponent, we obtain that $\Delta\sigma(T)$ follows the power law $\Delta\sigma \propto (T - T_{\mathrm{c}})^{-s}$ where $s = \nu(2 + z - d)$ [7]. Finally we obtain that

$$X^{-1} = \frac{1}{s}(T - T_{\mathrm{c}}). \tag{3.3}$$

The s exponent depends on the nature of the fluctuating system [7]. The main source of uncertainty of such analysis comes from the procedure of estimating σ_R. It may be large within the vicinity of the onset transition temperature. In order to avoid large uncertainties in this work we discuss only the data close to the temperature at which $\rho = 0$. This region reflects the mesoscopic granularity of the samples. Figure 4 presents examples of the temperature dependence of X^{-1} calculated for both studied samples, the exponents are collected in Table. The minimum on the high temperature side of the plots corresponds to the pairing transition temperature. When the temperature is decreased below T_1, the plot of X^{-1} versus temperature shows typical behaviour of granular superconductors [8]. First, it increases until a field dependent maximum is reached. Then, X^{-1} decreases in such a way that at temperature range close to the zero resistance the plots may be fitted to straight lines. In the case of the sample annealed at 830°C two regions of different slopes may be distinguished. In both samples, the width of the paracoherent state is large, it extends over several K. The exponents obtained

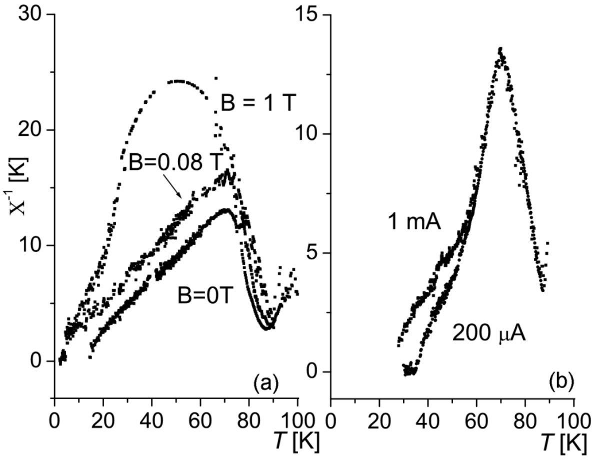

Fig. 4. Temperature dependence of X^{-1} calculated for the studied samples. (a) 830°C for 1 min, (b) 860°C, 2 min.

for the samples at low current density and at a very low magnetic field (4.2–4.4) are interpreted as an effect of critical fluctuations occurring before the coherence transition [7]. It corresponds to the dynamical critical exponent $z = 4.1$, which is considered as characteristic of a percolation-like transition in granular array in ceramic high-temperature superconductor (HTSC) [2, 8]. It has been observed in the samples with a wide transition, that is those with a large number of weak links between the superconducting grains. The physical nature of the transition is still under discussion. Models of spin glass, chiral glass, and gauge glass transitions have been considered as possible models of the coherence transition in granular superconductors [9].

Both the current and magnetic field is expected to increase the width of the paracoherent state. They do so indeed, but apart from that, another interesting effect of magnetic field on the superconducting transition may be observed. The field as small as $B = 0.125$ T causes a large change in the shape of X^{-1} maximum and increases the slope of its decrease towards the low temperatures (the s exponent corresponding to the transition in the magnetic field is about 2). The maximum becomes wide and rounded. The deviations from the power-law dependence in strong magnetic fields have been observed before [10], but they occurred in the field larger than 2 T. The effect observed in our sample probably reflects widening of the pairing transition in magnetic field. That is, the region below T_1 in which the thermal fluctuations occur widens in the magnetic field. The slope of the temperature dependence of X^{-1} in magnetic field is difficult to explain. The s exponent close to 2.5 has also been observed in $YBa_2Cu_3O_7$ in strong magnetic field [7]. It should be noted that all the granular high-T_c superconductors in which superconducting transition were studied before had much larger granule size (a few μm [2, 7, 8, 11] in comparison to 0.2 μm in our samples).

4. Conclusions

Even the samples containing only a few percent of a superconducting phase dispersed in a system of the small granules transit into a long-range ordered superconducting state.

Pairing transition temperature occurring in isolated granules depends only on the magnetic field. Coherence transition is very sensitive to the current flowing through the sample, magnetic field and the phase composition and microstructure of the sample.

The analysis of superconducting transition by means of the logarithmic derivative of paraconductivity shows the mesoscopic granularity of the samples. Unexpected influence of the magnetic field on the transition has been observed.

References

[1] *Superconducting Glass-Ceramics in Bi–Sr–Ca–Cu–O: Fabrication and Its Application*, Ed. Y. Abe, World Scientific, Singapore 1997.

[2] P. Pureur, R. Menegotto Costa, P. Rodrigues, Jr., J. Schaf, J.V. Kunzler, *Phys. Rev. B* **47**, 11420 (1993).

[3] M. Gazda, *Physica C* **411**, 170 (2004).

[4] X.Y. Lang, Q. Jiang, *Solid State Commun.* **134**, 797 (2005).

[5] J. Roa-Rojas, R. Menegotto Costa, P. Pureur, P. Prieto, *Phys. Rev. B* **61**, 12457 (2003).

[6] B. vom Hedt, K. Westerholt, *Physica C* **243**, 389 (1995).

[7] R.J. Joshi, R.B. Hallock, J.A. Taylor, *Phys. Rev. B* **55**, 9107 (1997).

[8] A.R. Jurelo, J.V. Kunzler, J. Schaf, P. Pureur, J. Rosenblatt, *Phys. Rev. B* **56**, 14815 (1997).

[9] E. Granato, *Physica C* **408-410**, 505 (2004).

[10] F.W. Fabris, J. Roa-Rojas, P. Pureur, *Physica C* **354**, 304 (2001).

[11] A.R. Jurelo, I. Abrego Castillo, J. Roa-Rojas, L.M. Ferreira, L. Ghivelder, P. Pureur, P. Rodrigues Jr., *Physica C* **311**, 133 (1999).