

# Magnetic Field Influence on the Superconducting Transition in Granular (BiPb)–Sr–Ca–Cu–O Superconductors

M. GAZDA AND T. KLIMCZUK

Faculty of Applied Physics and Mathematics, Gdańsk University of Technology

G. Narutowicza 11/12, 80-233 Gdańsk, Poland

In this work we present the studies of the influence of magnetic field on the superconducting transition in granular (Bi,Pb)–Sr–Ca–Cu–O superconductors. Special attention was devoted to the samples containing relatively small granules (smaller than 30 nm) of the 2212 superconducting phase. The samples were obtained by the glass-ceramic technology. The transition into the superconducting state was observed in the samples containing the 2212 phase in the form of crystallites with dimension equal or larger than 20 nm. Typical for granular superconductors, a two-stage character of the superconducting transition has been observed. Magnetic field influences the temperature dependence of resistivity of the samples containing granules of the 2212 phase in the superconducting phase, also in case of the samples for which no transition in the  $R(T)$  plot is observed. In the studied samples the magnetoresistance was positive in the whole range of magnetic fields.

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## 1. Introduction

A granular metal or superconductor is composed of metallic or superconducting grains embedded in an insulating matrix. Electronic conduction in the system of grains in the superconducting state is a complex phenomenon because both single electron and Cooper pairs tunnelling may occur. Apart from that, the Coulomb effects and various aspects of disorder influence the transport phenomena. So far, the experimental studies have given a variety of results. For example, Gerber et al. observed that the resistivity of a granular metal below critical temperature was much higher than in the normal state [1], while Chui et al. reported three types of temperature dependence of resistivity of granular aluminium: exponential,  $\propto T^{1/2}$  and logarithmic [2].

In this work we present the studies of the influence of magnetic field on the superconducting transition in granular (Bi,Pb)–Sr–Ca–Cu–O superconductors. Special attention was devoted to the samples containing relatively small granules (smaller than 30 nm) of the 2212 superconducting phase.

## 2. Experimental

The samples were produced by the solid state crystallization. First, the samples of  $(\text{Bi}_{0.8}\text{Pb}_{0.2})_4\text{Sr}_3\text{Ca}_3\text{Cu}_4\text{O}_x$  glass were prepared from reagent grade:  $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ ,  $\text{PbO}$ ,  $\text{CuO}$ ,  $\text{Sr}(\text{NO}_3)_2$  and  $\text{CaCO}_3$ . The substrates were mixed in the (Bi,Pb):Sr:Ca:Cu ratio 4:3:3:4 and calcinated at 820 °C for 10 h. Then, they were melted in a platinum crucible at 1250 °C for about 10 min, and

quenched. The glass was cut into bars of similar dimensions ( $2 \times 1 \times 8 \text{ mm}^3$ ) and polished before further thermal treatment. The crystallization was carried out in a tube furnace at temperature between 650 °C and 850 °C for 0.5–4 min. The phase composition and microstructure of the samples were checked by X-ray diffraction (XRD), atomic force microscopy (AFM) and/or scanning electron microscopy (SEM). The resistivity was measured between 2 K and 300 K in magnetic field up to 9 T.

## 3. Results and discussion

The XRD analysis showed that the metallic and superconducting phases formed as a result of a solid state crystallization of the glass were  $(\text{Bi,Pb})_2\text{Sr}_2\text{CaCu}_2\text{O}_x$  (2212) and  $(\text{Bi,Pb})_2\text{Sr}_2\text{CuO}_x$  (2201) [3]. The radius of oval grains of the 2212 and 2201 phases depends on the annealing conditions. It was about 30 nm and 10 nm in case of the samples annealed at 840 °C for 0.5 min and 650 °C for 4 min, respectively. The width of the region between two adjacent granules was about a few nm. The semiconducting or insulating matrix consisted of the amorphous material with a small amount of crystalline non-metallic phases (e.g.  $\text{CuO}$ ,  $\text{CaO}$ ,  $\text{Ca}_2\text{PbO}_4$ ) [3].

The magnetisation as a function of temperature of the samples crystallised at 650 °C for 2 min and 750 °C for 0.5 min is plotted in Fig. 1. It may be seen that both samples exhibit a weak but clear diamagnetic signal, with the transition temperature between 60 K and 70 K. It means that the samples contain the superconducting grains and it is the 2212 phase which may be associated with the transition. It should be noted that the superconducting

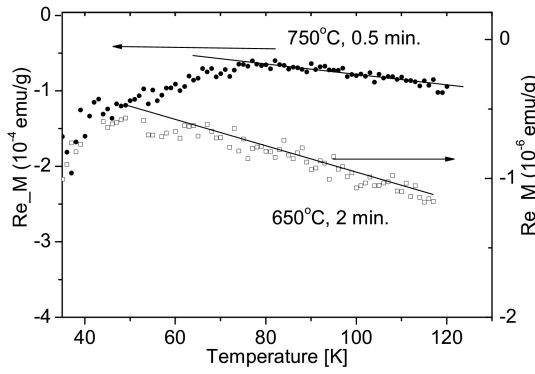


Fig. 1. Zero field cooling AC magnetization ( $H_{DC} = 10$  Oe,  $H_{AC} = 5$  Oe,  $f = 5$  kHz) as a function of temperature of the samples crystallised at  $650^\circ\text{C}$  for 2 min and  $750^\circ\text{C}$  for 0.5 min.

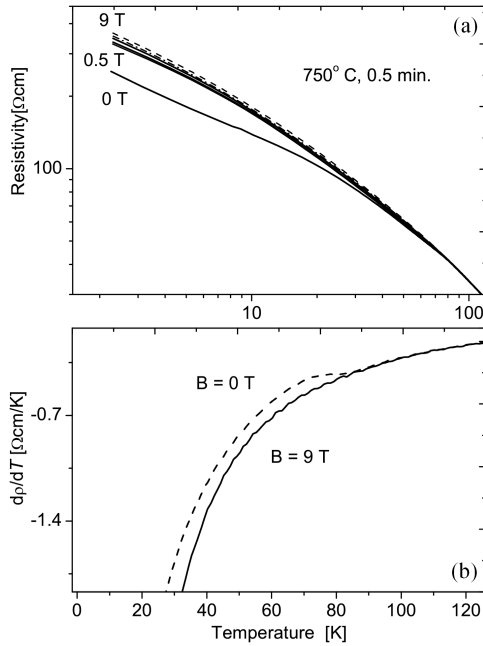


Fig. 2. Temperature dependence of resistivity (a) and temperature derivative of resistivity (b) of the sample annealed at  $750^\circ\text{C}$  for 0.5 min.

grains in these samples are isolated and the samples do not have the superconducting transition in the  $\rho(T)$  plot (Fig. 2).

The results of the measurements of resistivity of the samples annealed at  $750^\circ\text{C}$  and  $840^\circ\text{C}$  for 0.5 min, measured between 2 K and 120 K in a magnetic field below 9 T are presented in Figs. 2 and 3. The figures show also the temperature derivative of the resistivity plotted as a function of temperature. Several characteristic features common to both samples may be observed. The resistivity of the material in the normal state did not depend on the magnetic field. On the other hand, below the critical temperature of the 2212 phase the magnetic field changes

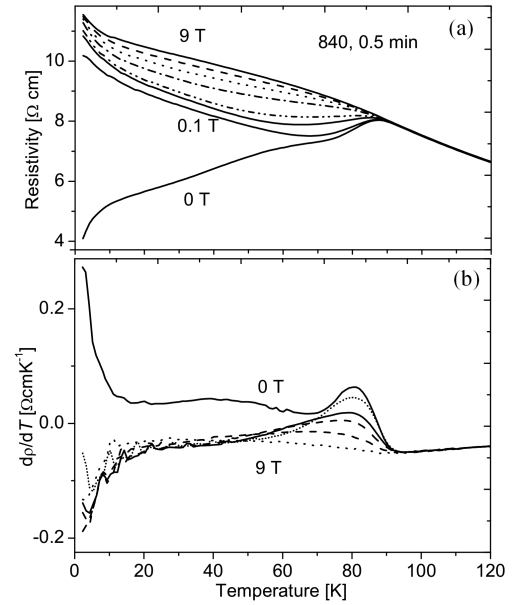


Fig. 3. Temperature dependence of resistivity (a) and temperature derivative of resistivity (b) of the sample annealed at  $840^\circ\text{C}$  for 0.5 min.

the resistivity also in the sample without the transition. In both cases, the magnetoresistance is positive. No super insulator behaviour reported by Gerber et al. [1] in a material consisting of isolated superconducting grains was observed. Figure 3 shows a typical feature for granular superconductors, a two-stage character of the superconducting transition. It should be noted that the low-temperature parts of the derivative plots, taken in the magnetic field (0.1–9 T) are similar one to another. It means that the intergrain superconducting coupling has been mostly destroyed even by a very weak field. On the other hand, a part of the 2212 grains remained in the superconducting state even at  $B = 9$  T.

#### 4. Conclusions

The 2212 granules even if there is no resistive superconducting transition, transit into the superconducting state below 70–60 K. In the samples with the resistive superconducting transition the magnetic field both hinders the tunnelling and destroys the Cooper pairs in the granules. The magnetic field of 9 T was smaller than the critical field of most of the studied samples.

#### References

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