

# Controlled Magnetoresistance in $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ –CuO Composites at 77 K

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Received January 28, 2003

**Abstract**—We have studied the low-temperature magnetoresistance of  $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$ –CuO composites obtained by fast sintering technique and established a relation between the probing to critical current density ratio  $j/j_c$  and the shape of the magnetoresistance curve  $\rho(H)$ . For  $j/j_c < 1$ , the electric resistance arises at a threshold value of the magnetic field strength  $H_c$ . For  $j/j_c \geq 1$ , a linear variation of  $\rho(H)$  at 77 K in the range from 0 to 14 Oe can be provided by selecting the CuO content (in the 15–30 vol % interval) and the  $j$  value (in the 0.003–0.2 A/cm<sup>2</sup> range). In the latter case, the slope  $d\rho/dH$  (i.e., the sensitivity of the electric resistivity with respect to the magnetic field) is 1–20 m $\Omega$  cm/Oe and the relative field-induced increase in the resistivity  $\rho_0 = (\rho(H) - \rho(H = 0))/\rho(H = 0)$  amounts to 1320 and 685% at  $H = 200$  and 35 Oe, respectively. Composites possessing controlled magnetoresistance are promising materials for the active elements of magnetic field sensors capable of operating at a practically convenient liquid nitrogen temperature. © 2003 MAIK “Nauka/Interperiodica”.

Previously [1], we presented preliminary data on the magnetoresistance of composites based on high-temperature superconductors (HTSCs). The composites, representing a system of artificial Josephson junctions with a non-HTSC component playing the role of barriers between HTSC grains, possess large magnetoresistance in a broad range of temperatures (below the superconducting temperature  $T_c$  of the HTSC component) and relatively weak (below 200 Oe) magnetic fields. This property allows such composites to be used in magnetic field sensors capable of operating at a practically convenient liquid nitrogen temperature.

The effect of magnetic field on the resistance of pure HTSC ceramics was studied shortly after the discovery of the phenomenon of high-temperature superconductivity [2–10]. A strong sensitivity of the resistivity  $\rho$  of HTSC materials to a weak external magnetic field is observed in a very narrow temperature interval below  $T_c$ . To obtain a significant response at  $T = 77$  K, it is necessary to employ high transport currents ( $\sim 10^2$ – $10^3$  A/cm<sup>2</sup>), which encounters considerable technical difficulties related to the heat removal from current-carrying leads [3, 5] and strong magnetic fields ( $H \sim 10$ – $60$  kOe). The magnetic field hysteresis of the electric resistance [4, 6, 7, 9–11] and the nonlinear character of the  $\rho(H)$  function [3–7, 11] also restrict the possible applications of magnetoresistive HTSC ceramics.

Below we report the results of a thorough investigation of the magnetoresistance  $\rho(H)$  at various transport current densities  $j$  in a series of composites based on yttrium ceramics and copper oxide. The parameters of the magnetoresistance observed at 77 K in these

HTSC–CuO composites show more optimistic prospects for the practical use of such materials in magnetic field sensors.

The samples of composites were synthesized from an HTSC composition  $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$  (referred to below as YBCO) and copper oxide CuO by fast sintering technique as described previously [1, 12]. The samples were annealed according to the following schedule: 2 min at 910°C, 3 h at 350°C, and cooling down to room temperature with the furnace. The X-ray diffraction measurements confirmed the absence of foreign phases in the final composites. We have synthesized and studied the samples of two different compositions denoted below as YBCO + 15 vol % CuO and YBCO + 30 vol % CuO (with an HTSC component fraction of 85 and 70 vol %, respectively).

The electric resistivity as a function of the temperature,  $\rho(T)$ , and the magnetic field strength,  $\rho(H)$ , was studied by a standard four probe technique using samples with typical dimensions of  $1 \times 2 \times 12$  mm. The distance between the potential contacts was  $\sim 10$  mm and the probing (transport) current density was varied in the range from 0.002 to 0.2 A/cm<sup>2</sup> (which corresponds to a total current of  $\sim 0.1$ – $10$  mA). The superconducting state ( $R = 0$ ) was detected to within  $\sim 10^{-6}$   $\Omega$  cm. The critical current density  $j_c$  was determined using the initial part of the current–voltage characteristic according to the conventional criterion of  $10^{-6}$  V/cm [13]. During the magnetoresistance measurements, the magnetic field was applied perpendicularly to the electric current direction. No special measures were taken for shielding the Earth’s magnetic field. The magnetization measure-

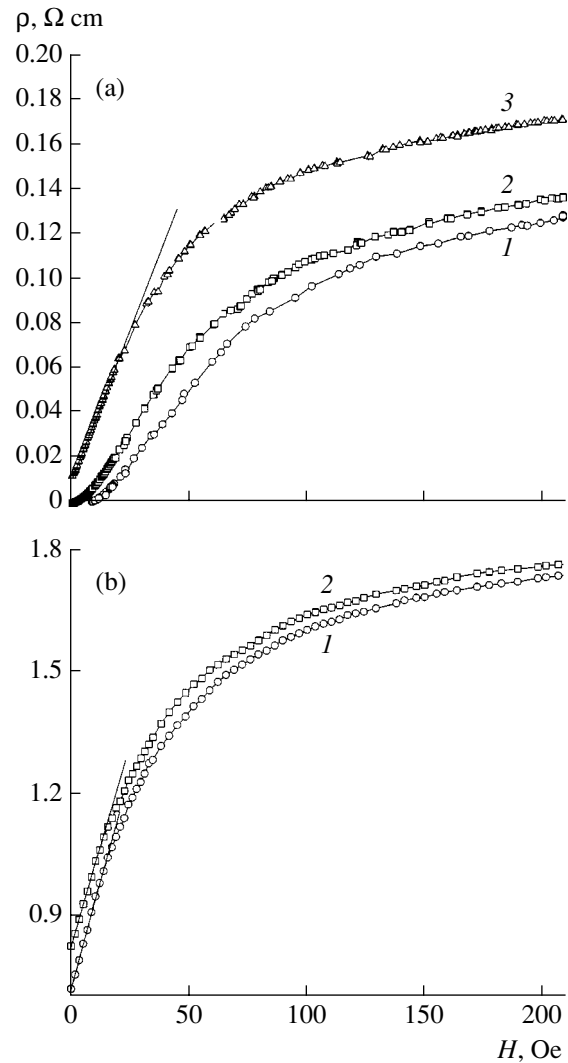
ments were performed using a vibrating-sample magnetometer [14].

Figure 1 shows the experimental  $\rho(T)$  curves of the YBCO + 15 vol % CuO and YBCO + 30 vol % CuO composites measured at 77 K using different values of the probing transport current. All variants of the observed  $\rho(H)$  curves can be divided into three types depending on the probing current density (at a given temperature). When the current density is below the critical value,  $j < j_c$ , there is an interval in which  $\rho \leq 10^{-6} \Omega \text{ cm}$ . Only beginning with a certain  $H_c$  ( $H_c \approx 10 \text{ Oe}$  for YBCO + 15 vol % CuO at  $j = 0.12 \text{ A/cm}^2$  and  $T = 77 \text{ K}$ ), for which the given current density is critical, does the resistivity exhibit a nonlinear growth with the magnetic field strength (Fig. 1a, curve 1). For  $j \approx j_c$ , the growth of  $\rho(H)$  begins at the origin of coordinates (Fig. 1a, curve 2). Finally, for  $j > j_c$ , the resistivity varies from a certain zero-field value  $\rho(H = 0)$  (Fig. 1, curve 3) and  $\rho(H)$  is a linear function of the field strength in the interval from 0 to 14 Oe with a slope of  $d\rho/dH \approx 2.5 \text{ m}\Omega \text{ cm/Oe}$ .

For the YBCO + 30 vol % CuO composition, the critical current  $j_c$  (77 K) is below  $0.001 \text{ A/cm}^2$  and all the  $\rho(H)$  curves measured in the entire interval of probing currents  $j$  (Fig. 1b) are similar to curve 3 in Fig. 1a. These  $\rho(H)$  plots are also linear in the interval from 0 to 14 Oe, but the slope is  $d\rho/dH \approx 17.5 \text{ m}\Omega \text{ cm/Oe}$ .

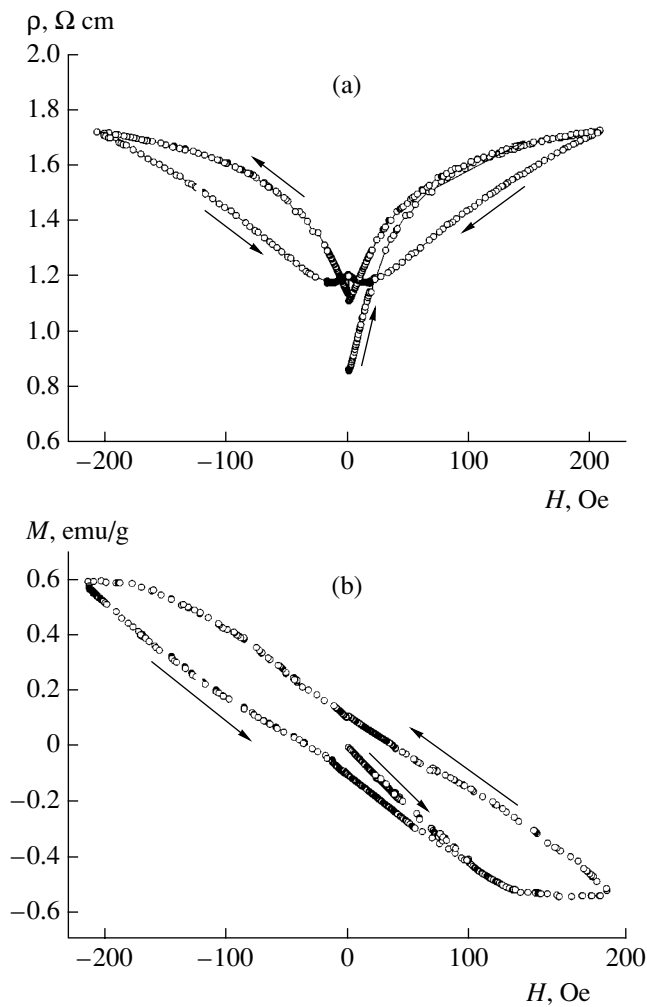
In the magnetic fields with  $|H| \leq 37 \text{ Oe}$ , both composites were characterized by coinciding  $\rho(H)$  curves measured when increasing and decreasing the absolute value of the field strength. For greater magnetic field amplitudes, the  $\rho(H)$  curves exhibit a hysteresis. Figure 2 shows the pattern of  $\rho(H)$  and the corresponding magnetization curve  $M(H)$  measured for the same specimen of YBCO + 30 vol % CuO with the field varied in the interval  $-210 \text{ Oe} \leq H \leq 210 \text{ Oe}$ . Note that the behavior of  $M(H)$  is typical of HTSC ceramics [11]. The  $\rho(H)$  loops are well reproduced in the course of multiply repeated field cycling. The thermomagnetic history can be reset by heating a sample above  $T_c$  followed by cooling in a zero field. A comparison of the  $\rho(H)$  and  $M(H)$  curves shows that the sample resistance is a complicated function of the magnetization.

The results of  $\rho(H, j)$  measurements at other temperatures were similar to those presented in Figs. 1 and 2. The ratio of the probing to critical current density,  $j/j_c$ , is the main parameter determining the type of the  $\rho(H)$  curve. Figure 3 shows the temperature dependence of the critical current density  $j_c(T)$  in the vicinity of  $T_c$ . Using these data and selecting the probing current density, it is possible to predict the shape of the  $\rho(H)$  characteristic of a given magnetic field sensor for any temperature in the range studied. In ceramics with natural grain boundaries, the values of  $j_c(77 \text{ K})$  range within  $10$ – $250 \text{ A/cm}^2$  [5, 6, 15]. For this reason,  $\rho(H)$  curves of various types such as 1–3 in Fig. 1a are more difficult to obtain in pure ceramics than in composites.

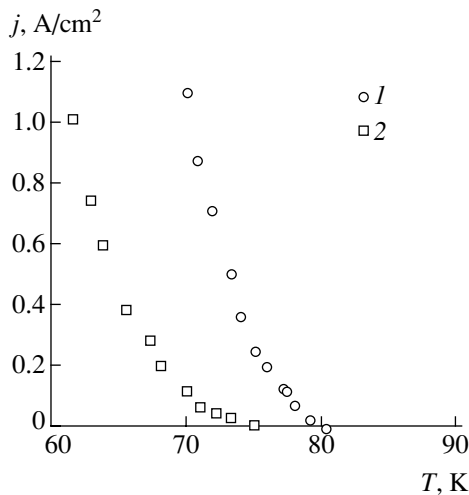


**Fig. 1.** Plots of the resistivity  $\rho$  versus magnetic field strength  $H$  at  $T = 77 \text{ K}$ : (a) YBCO + 15 vol % CuO composite,  $j = 0.037 \text{ A/cm}^2 < j_c$  (1),  $j = 0.12 \text{ A/cm}^2 \approx j_c$  (2), and  $j = 0.37 \text{ A/cm}^2 > j_c$  (3); (b) YBCO + 30 vol % CuO composite,  $j = 0.0032$  (1) and  $0.032 \text{ A/cm}^2$  (2).

The phenomenon of magnetoresistance observed in polycrystalline HTSCs (including composites) is explained by the fact that these materials represent a system of Josephson junctions. The boundaries between the HTSC grains are weak bonds of the Josephson type, the electric resistance of which is highly sensitive to external magnetic fields [13]. The number of weak bonds per unit length is very large, on the order of  $10^3$  per mm for a typical size of superconducting granules reaching  $\sim 1.5 \mu\text{m}$  (according to scanning electron microscopy data). Accordingly, the response of one HTSC-grain boundary-HTSC junction has to be multiplied by this number. In the YBCO-CuO composites studied, copper oxide forms dielectric layers between HTSC grains [12]. By selecting the volume content of components, it is possible to control the



**Fig. 2.** Plots of (a) the resistivity  $\rho$  ( $j = 0.0032 \text{ A/cm}^2$ ) and (b) magnetization  $M$  versus magnetic field strength  $H$  for an YBCO + 30 vol % CuO composite at  $T = 77 \text{ K}$ . Arrows indicate the direction of field variation.



**Fig. 3.** Plots of the critical current density  $j_c$  versus temperature for (1) YBCO + 15 vol % CuO and (2) YBCO + 30 vol % CuO composites.

“strength” of the Josephson bonds so as to provide that the overall superconducting transition takes place at a preset temperature (in our case,  $\sim 80 \text{ K}$  for YBCO + 15 vol % CuO and  $\sim 76 \text{ K}$  for YBCO + 30 vol % CuO). Then, a significant magnetoresistance at  $77 \text{ K}$  can be observed for relatively weak magnetic fields and low probing current densities. In addition, CuO can be considered as an insulator at low ( $< 100 \text{ K}$ ) temperatures [16]. This leads to an increase in the resistivity of YBCO–CuO composites in the normal state as compared to that of the pure HTSC ceramics [12]. For this reason, the slope  $d\rho/dH$  (i.e., the sensitivity with respect to the magnetic field) obtained in the composites ( $2\text{--}20 \text{ m}\Omega \text{ cm/Oe}$ ) is greater by at least two orders of magnitude than that in polycrystalline HTSCs ( $0.01\text{--}0.15 \text{ m}\Omega \text{ cm/Oe}$ , as estimated from the data reported in [2, 3, 5, 6]).

Another important parameter characterizing the magnetoresistance is the relative field-induced increase in the resistivity  $\rho_0 = (\rho(H) - \rho(H = 0))/\rho(H = 0)$ , which is the factor by which the resistivity grows upon application of the magnetic field  $H$ . Of course, the growth in the resistivity relative to that in the superconducting state ( $R = 0$ ) is always very large. In practice, it is more important to estimate the resistivity growth relative to a certain finite value of  $\rho(H = 0)$ . In YBCO + 15 vol % CuO, the value of  $\rho_0$  for  $j > j_c$  (Fig. 1a, curve 3) amounts to 1320% at  $H = 200$  and 685% at  $35 \text{ Oe}$  (i.e., in the region of reversibility). The zero-field resistivity  $\rho(H = 0, T = 77 \text{ K})$  at  $j = 0.37 \text{ A/cm}^2$  is  $8 \text{ m}\Omega \text{ cm}$ . In YBCO + 30 vol % CuO, the value of  $\rho_0$  for  $j = 0.0032 \text{ A/cm}^2$  amounts to 140 and 78% at  $H = 200$  and  $35 \text{ Oe}$ , respectively, and  $\rho(H = 0, T = 77 \text{ K})$  is  $725 \text{ m}\Omega \text{ cm}$ .

The behavior of  $\rho(H)$  such as that depicted in Fig. 1a can be used in devices intended to signal in response to a threshold magnetic field strength  $H_c$ . HTSCs materials operating in this regime are called supermagnetoresistors [5]. In our composites, the required  $H_c$  value can be adjusted by selecting the volume content of a non-HTSC component and the transport current density.

Thus, the behavior of  $\rho(H)$  in the composites under study is determined by the probing (transport) to critical current density ratio  $j/j_c$ . For  $j/j_c$ , the electric resistance arises at a threshold value of the magnetic field strength  $H_c$ . For  $j/j_c \geq 1$ , there is a region in which  $\rho(H)$  is a linear function with a high value of the sensitivity with respect to the field strength:  $d\rho/dH \sim 1\text{--}20 \text{ m}\Omega \text{ cm/Oe}$ . The relative increase in the resistivity  $\rho_0$  in the region of reversibility at  $T = 77 \text{ K}$  reaches several hundred percent. The resistivity response to magnetic field in the  $\text{Y}_{3/4}\text{Lu}_{1/4}\text{Ba}_2\text{Cu}_3\text{O}_7\text{--CuO}$  composites studied (at relatively low values of the probing current,  $j \sim 1 \text{ mA/cm}^2$ ) is two orders of magnitude greater than that in pure polycrystalline HTSC ceramics. All these results are indicative of good prospects for using these composites in the active elements of magnetic field sen-

sors capable of operating at a practically convenient liquid nitrogen temperature. In the region of reversible magnetoresistance ( $|H| \leq 37$  Oe), the characteristics of such sensors ( $\rho_0$ ,  $d\rho/dH$ ,  $\rho(H)$ ) are comparable with those of the best devices based on magnesium oxide [17].

**Acknowledgments.** The authors are grateful to A.F. Bovina for the X-ray diffraction analysis of composites, to A.D. Balaev for his help in conducting magnetic measurements, to D.M. Gokhfeld for his help in work and discussion of results, and to S.V. Komogortsev for fruitful discussions.

This study was supported in part by the Joint Program "Yenisei" of the Krasnoyarsk Regional Science Foundation and the Russian Foundation for Basic Research (project no 02-02-97711) and by the Siberian Division of the Russian Academy of Sciences within the framework of the Lavrentiev Competition of Young Scientist Projects 2002.

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*Translated by P. Pozdeev*