Magnetic Field Dependence of Intergrain Pinning Potential in Bulk Granular Composites YBCO + CuO Demonstrating Large Magneto-Resistive Effect

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Abstract The broadening of the resistive transition in magnetic field and isotherms of magnetoresistance of bulk composites Y–Ba–Cu–O + CuO have been studied. These composites exhibit large magneto-resistive effect in a wide temperature range below T_C due to weakening of Josephson coupling in this system. The broadening of the resistive transition and magnetoresistance are explained well by the Ambegaokar–Halperin (AH) model for phase slip in Josephson junctions. The magnetic field dependence of pinning potential in the intergrain boundaries deduced from AH model found out to be similar to that of critical current of an array of Josephson junctions. The values of pinning energy point out that the large magneto-resistive effect observed in the composites results from flux flow-like processes at the intergrain boundaries.

Keywords Bulk granular High- T_C superconductor · Magneto-resistive effect · Josephson junction network · Inter-grain pinning potential

1 Introduction

The Ambegaokar–Halperin (AH) model [1] of phase slip in Josephson junction have been successfully applied for description of broadening of resistive transition of polycrystalline High- T_C superconductors (HTSCs) [2–7] in magnetic field. The AH model describes the time-averaged voltage produced by a thermal-noise current superimposed on

D.A. Balaev (⊠) · A.A. Dubrovskiy · S.I. Popkov · K.A. Shaykhutdinov · M.I. Petrov Kirensky Institute of Physics of RAS, 660036, Krasnoyarsk, Russia e-mail: smp@iph.krasn.ru a current-driven Josephson junction. Tinkham [8] and Tinkham and Lobb [9] have argued that AH model can be applied for description of dissipation in granular superconductors in a magnetic field. Periodic potential considered in AH model is a good mathematical equivalent of pinning sites in network of Josephson junctions realized on intergrain boundaries of granular HTSCs. Jumps of flux vortices from site to site lead to phase slip on 2π which results in dissipation. The Josephson coupling energy $E_J = \hbar I_C / e$ (I_C —is the critical current in absence of fluctuations) is unambiguously associated with the pinning energy U_p . The magnetic field dependence of pinning potential derived from treatment of experiments with AH model can be approximated by power law function [3-6]. However, in most of experiments [3–7] the range of small magnetic fields ($\sim 2 \div 20$ Oe) was not studied. Nevertheless, the $U_p(H)$ dependence may exhibit a peculiarity in this range [10] so far the $U_p(H)$ dependence should behave likely magnetic field dependence of critical current of network of Josephson junctions (which determine transport properties of granular HTSC). To our best knowledge, only in [10] the $U_p(H)$ dependence was deduced from magnetoresistance R(H) measurements of polycrystalline Bi-Pb-Sr-Ca-Cu-O and this dependence correlates with averaged Fraunhofer function describing the magnetic field dependence of Josephson current of array of Josephson junctions [11]. On author's opinion, the behavior of both the resistive transition and magnetoresistance in materials based on Y-Ba-Cu-O ceramic in the range of small magnetic fields would be studied and corresponding field dependence of pinning potential would be scrutinized.

In this paper, we report in detail the measurements of R(T) and R(H) dependences of polycrystalline compos-

ite samples YBCO + CuO in the range of magnetic fields¹ $\sim 2 \div 20$ Oe and apply the AH model to deduce the $U_p(H)$ dependence and to recognize the character (flux creep, flux flow) of dissipation processes to take place in these materials.

This study has a partial goal so far such bulk twophases composite materials have been found to exhibit large magneto-resistive effect in a wide temperature range (\sim 50÷ 90 K) for weak (less than 100 Oe) magnetic fields and the resistivity of these composites is highly sensitive to the external magnetic field [12]. This effect may be used in magnetic field sensor devices operating at cryogenic temperatures (for example, 77.4 K) [12, 13]. Also, the large MR effect was found in bulk polycrystalline $Bi2212 + USr_2CaO_6$ [14, 15] and Bi2212 + MgO composites [16] and granular YBCO thick films [17] and BSCCO films [18]. In the composites, the nonsuperconducting component forms boundaries between the HTSC crystallites and these composites may be considered as "model" granular HTSCs with reduced Josephson coupling [4, 6, 12]. More thorough understanding of physical mechanisms responsible for the large magnetoresistance of HTSC based composite materials is necessary.

2 Experimental

The composite samples containing 85% Vol. of Y_{3/4}Lu_{1/4}- $Ba_2Cu_3O_7$ and 15% Vol. CuO (denoted as YBCO + 15CuO) and 70% Vol. of Y_{3/4}Lu_{1/4}Ba₂Cu₃O₇ and 30% Vol. CuO (YBCO + 30CuO) are the same ones as in our previous works [12, 13]. The preparation technique has been described in [12]. Magnetic measurements revealed a single superconducting phase with the critical temperature $T_C = 93.5$ K, which is similar to pure $Y_{3/4}Lu_{1/4}Ba_2Cu_3O_7$. The R(H, T) = V(H, T)/I (V—is the voltage drop on the sample) dependences have been measured by the standard four-probe technique. The critical current I_C was determined from initial part of I-V characteristics by the standard 1 μ V/cm criterion. The magnitude of I_C for sample YBCO + 15CuO is 1.5 mA at 77.4 K and 100 mA at 4.2 K. The composite YBCO + 30CuO has no critical current at 77.4 K; the value $I_C(4.2 \text{ K})$ 30 mA for this sample. The resistivity in the normal state (at 95 K) is ~ 0.7 cm and \sim 3.0 cm for samples YBCO + 15CuO and YBCO + 30CuO, respectively. The transport current was perpendicular to the magnetic field direction. The samples were cooled in zero magnetic field with no screening in the earth's magnetic field.

3 Results and Discussion

Figure 1 shows the resistive transition of composite YBCO + 15CuO in zero magnetic field and magnetic fields 2, 5, 10, 15, 20 Oe measured at transport current I = 1 mA. (Resistive transition of sample YBCO + 30CuO at H = 0 is shown in insert of Fig. 1).

The R(T) curves exhibit two-step transition typical for granular HTSCs [2–7, 12, 13, 19]. A sharp drop of resistance at $T_C = 93.5$ K is related to the superconducting transition in the HTSC crystallites. The second, smooth part of R(T) reflects the superconducting transition in the network of Josephson junctions realized in the composites. Namely, this part of R(T) is very sensitive to weak magnetic fields. The R(T) dependences in Fig. 1 are normalized to the value of resistance of this smooth part R_{NJ} , i.e., to R (91 K). Magnetoresistance curves of sample YBCO + 15CuO at 77 K (I = 1 mA) and sample YBCO + 30CuO (I = 1 mA) at various temperatures are shown on Fig. 2.

At least two theoretical models account for magnetoresistance may be considered. The first one is the classical Anderson theory of flux creep or flux flow processes [20]. In the Anderson theory, the magnetoresistance caused movement of vortices defines as:

$$R = R_0 \exp(-U_p(H, T)/k_B T)$$
⁽¹⁾

where R_0 —is the normal resistance, $U_p(H, T)$ —is the effective magnetic field and temperature dependence of pinning energy, k_B —is the Boltzmann constant. For the case of classical flux creep [21], where $U_p \gg k_B T$, the pinning



Fig. 1 Temperature dependences of resistance normalized to the value R_{NJ} (the normal resistance of intergrain boundaries) for the YBCO + 15CuO composite measured at various magnetic fields H (I = 1 mA). In the *insert*: the resistive transition for YBCO + 15CuO composite at H = 0, I = 1 mA. *Solid curves*—results of best by AH model

¹In our previous study [6], the R(T) dependences of composites YBCO + CuO have been measured and analyzed within AH and flux creep models for magnetic fields above 40 Oe while the magnetoresistance in the range of interest (2 ÷ 20 Oe) was not studied.



Fig. 2 The magnetoresistance R(H) dependences for the composites measured at various temperatures (I = 1 mA). Solid curves—results of best fits with AH model

energy should not depend on temperature and the resistance behaves as:

$$R \sim \exp(-U_p/k_B T). \tag{2}$$

Behavior predicted by (2) is observed in the vicinity of transition to "zero resistance state." When the experimental data presented on the Fig. 1 are plotted in the coordinates $\lg R$, 1/T, these dependences found out to be strongly non-linear. We have shown in [6] that for the composites YBCO + CuO the dependence (2) takes place only in the field range above \sim 1 kOe. The experimental R(T) data were treated with (1) to deduce the temperature dependence of pinning potential as:

$$U \sim (1 - T/T_C)^n \tag{3}$$

[2–5, 7, 8]. We have found a strong dependence of exponent n vs. $H: n \approx 2.2$ at H = 0; n = 1.5 at H = 20 Oe; the n value tends to ~ 1 at high fields. So, the fitting procedure of R(T) with (1) involves two field dependent variable fitting parameters: U(H, at T = const) and n in (3), i.e., temperature dependence of pinning potential.

From the other hand, the R(T) data may be fitted well with the AH model [1]. According to [1], in the limit of small transport current the R(T) dependence at $T < T_C$ defines as:

$$R = R_N \{ I_0(\gamma/2) \}^{-2}, \tag{4}$$

where R_N —is the resistance in the normal state, I_0 —is the modified Bessel function, γ —is the ratio of the Josephson coupling energy E_J to thermal energy. In the spirit of ideas of Tinkham [8, 9], the Josephson coupling energy may be considered as pinning energy, therefore, $\gamma = E_J(T)/kT =$ $U_p(T)/kT$. Equation (4) has in it both flux creep ($\gamma \gg 1$)





Fig. 3 The parameter γ , deduced from results of the best fits of R(T) dependences by AH model—symbols (*closed circles*—results of this study, *open circles*—results of work [6] for the same sample). *Curves*— $\gamma(H) = U_p(H)/kT$ deduced from results of best fits of R(H) dependences by AH model (Fig. 2). Every $\gamma(H)$ data is related to the experimental temperature

and flux flow regimes (when thermal energies become comparable to the barrier heights [21]) and reproduces the Slike shape of resistive transition in magnetic fields. Some researchers [2, 4, 5] used dependence of $E_I(T)$ (i.e., $U_p(T)$) in the form (3). But for the composites with insulator (which is the object under study) the network of tunnel-like type Josephson junctions determines its transport properties [12]. So, it is necessary to use the $E_J(T)$ dependence which reflects Josephson tunneling through insulating barriers. We have found that using of classical Ambegaokar-Baratoff (A–B) dependence of $J_c(T)$ [22] as $E_J(T)$ in (4) allows to describe successfully the resistive transitions of composites YBCO + CuO. Solid curves in Fig. 1 are results of the best fit by AH model with A–B dependence of $E_I(T)$ (i.e., $U_p(T)$) for magnetic field range $H \leq 20$ Oe. The R_N value is also the same for all fields and it corresponds to R_{NJ} (see Fig. 1). So, the fitting procedure with AH expression has only one adjustable parameter—the value of $U_p(T=0)$ at various H, while similar fit with flux creep relation (2) requires two field-dependent parameters: $U_p(T=0)$ and n in (3). We consider this fact as an additional proof that namely AH mechanism is account for the dissipation in the low magnetic field range.

The field dependence of pinning energy U_p obtained from results of the best fit of the data of the Fig. 2 is shown in the Fig. 3 in double-logarithmic scale. The $U_p(H)$ data are presented on the Fig. 3 as the ratio of pinning energy and thermal energy, i.e., $U_p(T = 0) \times f_{AB}(T)/kT$ (where $f_{AB}(T)$ —is the normalized A–B temperature dependence). The circles are the data obtained for sample YBCO + 15CuO in this work and triangles are the data from [6] for the same sample. It is seen from Fig. 3 that the $U_p(H)$ dependence is not a simple power law function as it was concluded by many authors for polycrystalline HTSCs studied in magnetic field range above tens of Oersteds [2–6]. To clarify the functional dependence of $U_p(H)$, it is advisable to treat the R(H) curves by expression following from the AH model.

To fit the R(H) curves shown on the Fig. 2, besides parameter R_{NJ} , which to be held the same for various temperatures, it is necessary to know the functional dependence of $U_p(H)$. This dependence should be some drop-down curve which reflects the field dependence of I_C of Josephson junction network realized in polycrystalline HTSC (so far as $U_p \sim E_J \sim I_C$). We took the $U_p(H)$ in the form:

$$U_p(H) = \frac{U_p(H=0,T) - C_1}{H + C_2} \times C_2 + C_1,$$
(5)

where C_1 and C_2 are constants. Equation (5) approximates the averaged Fraunhofer function describing the magnetic field dependence of Josephson current of array of Josephson junctions [11]. A good agreement of experimental and theoretical R(H) curves is obtained using (4) with U_p determined by (5). $U_p(H = 0)$, C_1 and C_2 were fitting parameters (the U_p values at various temperatures were calculated taking into account $f_{AB}(T)$ temperature dependence). The Fig. 2 illustrates results of these fits. Deduced $U_p(H)$ dependences are plotted in the Fig. 3.

The kink on the $U_p(H)$ dependences is seen from the Fig. 3. About the point $H \sim 20$ Oe the dependence $\lg U_p$ vs. $\lg H$ changes its curvature. This behavior is seen from two independent types of fit: R(T) at various fields (symbols in the Fig. 3) and R(H). Similar shape of U_n was obtained in [10] on polycrystalline Bi-Pb-Sr-Ca-Cu-O system. Such peculiarity is specific for Josephson junction array [11]. In principle, it is possible to distinguish the field range where the dependence (5) may be approximated by the power law function. For example, in the range 20 Oe $\leq H \leq 2 \times 10^2$ Oe the $U_p(H)$ data on the Fig. 3 can be described well by $\sim H^{-0.39}$ function. Experimentally measured field dependences of critical current $I_C(H)$ of granular HTSCs also have similar form [11, 23]. The $I_C(H)$ dependences for the composite YBCO + 15CuO at various temperatures are shown on Fig. 4. Note that experimentally observed critical current is affected by thermal fluctuations [1] and the voltage criterion, therefore, fully coincidence of experimental $I_C(H)$ and $U_p(H)$ deduced with AH model is not expected.

Let us pay attention to the values of U_p deduced from R(T, H) fits. For sample YBCO + 15CuO at T = 77.4 K, I = 1 mA, and small H, the γ value is about 10 at $H \approx 0$. So, the dissipation in external magnetic fields about several Oersteds and current less than experimentally measured critical one (I = 1 mA, $I_C \approx 1.5$ mA) may be considered as flux creep process. Sample YBCO + 30CuO has no critical current at 77.4 K and the case $I > I_C$ takes place for all R(H)



Fig. 4 Experimental dependences of critical current (semi-log. scale) vs. external magnetic field for the composite YBCO + 15CuO at various temperatures

and $U_p(H)$ curves shown in Figs. 2 and 3 for this sample. It is seen that for this case the pinning energy becomes comparable to the thermal energy, which allows to characterize the dissipation regime as close to flux flow. This correlates with results of [13] where the magnetoresistance as a function of angle Θ (where $\Theta = \angle I$, **H**) was studied on the same composites. $\sin^2 \Theta$ dependence of magnetoresistance at $I > I_C$ was observed [13] which points out that the flux flow mechanism is responsible for the magneto-resistive effect in these materials. In this work, we deduced pinning energies at the intergrain boundaries of YBCO based composites and supported the conclusion of [13] that the magnetoresistance of these materials at weak magnetic fields is caused by flux flow-like processes.

4 Conclusions

Thus, in this work, the broadening on resistive transition of bulk YBCO + CuO composites in external magnetic field have been studied with the purpose to clarify the magnetic field dependence of pinning potential $U_p(H)$ at the intergrain boundaries. The AH model explains adequately experimentally measured R(T) and R(H) dependences. The $U_p(H)$ dependences deduced from treatment of experimental data within AH model are found to behave similarly to the field dependence of Josephson current of array of Josephson junctions. The large magneto-resistive effect observed in the HTSC based composites is caused by flux flowlike processes at the boundaries between superconducting crystallites.

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