Physica C 495 (2013) 39–43

Contents lists available at ScienceDirect

Physica C

journal homepage: www.elsevier.com/locate/physc

Magnetoresistance anomalies in ultra-thin granular $YBa_2Cu_3O_{7-\delta}$ bridges

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ARTICLE INFO

Article history:
Received 17 July 2013
Accepted 25 July 2013
Available online 7 August 2013

Keywords:
Magnetoresistance
Granular superconductors
$YBa_2Cu_3O_{7-\delta}$ bridges

ABSTRACT

We report on magnetoresistance measurements in 10 nm thick and submicron-wide granular $YBa_2Cu_3O_{7-\delta}$ bridges. The results show a strong dependence of the resistance on the magnetic field at low fields crossing over to a relatively weak field dependence at high fields. The field derivative of the resistance at high fields decreases as the temperature is lowered and eventually changes sign, exhibiting a *negative* slope in a wide field range in the Tesla regime. This negative slope is sensitive to the bias current, turning to be positive as the bias current increases. This complex magnetoresistance behavior is attributed to both phase slips in a distribution of strongly and weakly linked superconducting grains, and tunneling of quasiparticles between grains. The latter dominates at low temperatures and high fields, giving rise to the negative magnetoresistance slope.

1. Introduction

Extensive studies of superconductors with confined geometries revealed unusual phenomena that are normally absent in bulk or films of the material. These include magnetoresistance oscillations observed well below the transition temperature as first reported for granular Sn nano-wires [1] and a low-field *negative* magnetoresistance first reported for Al and Pb nano-wires [2,3]. Also, an intriguing phenomenon of *negative* rate of change of the magnetoresistance with field in the Tesla regime was recently observed in homogeneously disordered amorphous Pb ultrathin films [4], in tungsten-based nanowires and in superconducting ultrathin TiN networks [5]. Most of the publications related to the above phenomena describe measurements in conventional, low-T_c superconducting materials. In this paper we describe magnetoresistance measurements in ultra-thin submicron-wide $YBa_2Cu_3O_{7-\delta}$ (YBCO) granular bridges which exhibit the anomalies listed above, including magnetoresistance oscillations, negative magnetoresistance at low fields and negative magnetoresistance slope over a wide field range in the Tesla regime. Previously, we reported on the first two phenomena in a narrow YBCO bridge and discussed them in detail [6]. Here we focus on the complex behavior of the magnetoresistance background on which the oscillations are superimposed, in particular on the negative magnetoresistance slope observed in a wide field range in the Tesla regime. This negative slope is sensitive to the bias current, turning to be positive as the bias current increases. The complex behavior of the magnetoresistance background is attributed to both phase slips in a distribution of strongly and weakly linked superconducting grains, and tunneling of quasiparticles between grains. The latter dominates at low temperatures, giving rise to the negative magnetoresistance slope.

2. Experimental

Pulsed Laser Deposition (PLD) was used to grow 10 nm thick optimally doped epitaxial *c*-axis-oriented granular $YBa_2Cu_3O_{7-\delta}$ film on $10 \times 10\ mm^2$ (1 0 0) $SrTiO_3$ wafers. The atomic force microscope (AFM) image of the film shown in Fig. 1a demonstrates that the film is made of loosely connected grains of lateral size 30–100 nm. The film was spin-coated with poly(methyl methacrylate) (PMMA) electron-beam resist, and, subsequently, was baked for 90 s on a hotplate at 180 °C. A pattern of a 700 nm long and 80–500 nm wide bridges, including the contact-pads for the four-probe measurements, were written in the PMMA layer using a CRESTEC CABLE-9000C high-resolution electron-beam lithography system. Following development of the exposed PMMA using methyl isobutyl ketone (MIBK), and a standard argon ion milling process, the desired pattern was obtained. AFM image of a typical bridge and a line profile across the bridge are shown in Fig. 1b and c, respectively.

A Quantum Design Physical Properties Measurement System (model 6000) was exploited to measure the bridge resistance between 10 and 300 K, using DC bias currents between 0.25 and 20 μA. The magnetoresistance was measured in fields between

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http://dx.doi.org/10.1016/j.physc.2013.07.012

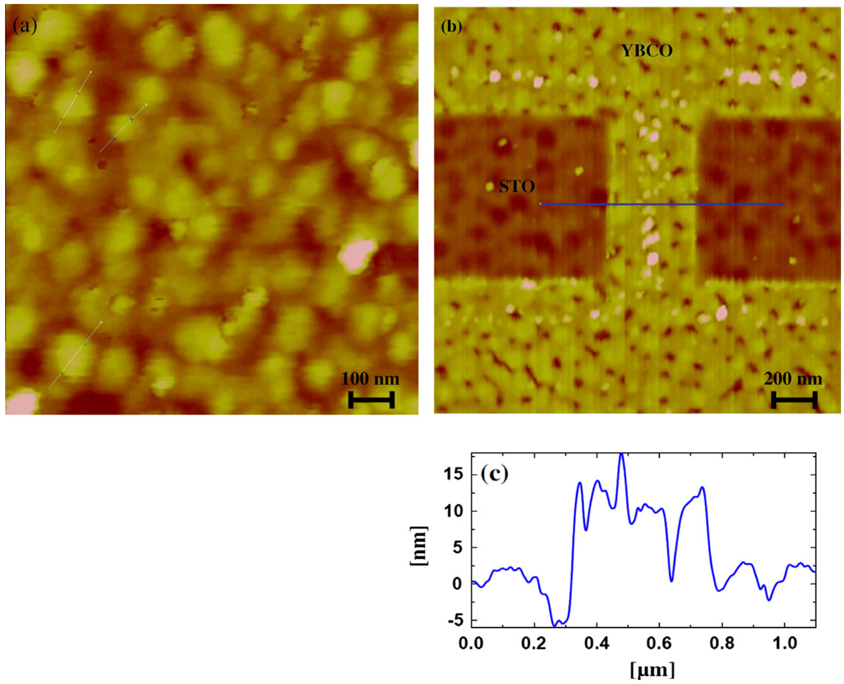

Fig. 1. (Colored in the web version) (a) AFM image of a 1×1 μm^2 area of the $YBa_2Cu_3O_{7-\delta}$ granular film. Bright colors represent higher areas and the brown areas are voids. The pink color represents areas with remnant of the PMMA resist. (b) AFM image of a 2×2 μm^2 area of the film, showing a patterned bridge with dimensions 500×700 nm^2. (c) A typical line profile across the bridge.

−5 and 5 T applied normal to the film surface (parallel to the *c*-crystallographic axis), keeping the temperature constant (with a stability of a few mK), in the range 20–80 K.

3. Results

Fig. 2 shows the temperature dependence of the resistance of a 500 nm wide bridge measured in various external fields. At zero field the onset of superconductivity is around 89 K reflecting the transition of the grains to a superconducting state. A transition broadening with field is apparent. Remarkably, some of these curves cross each other at low temperatures as demonstrated in the inset to the figure for 3 and 5 T. This unusual effect also manifest itself as a negative slope of the magnetoresistance as will be described later.

Fig. 3 summarizes in a semi-log plot the results of magnetoresistance measurements between 30 and 86 K in a field range ±5 T, using a bias current of 2 μA. The figure shows distinctive behavior of the magnetoresistance in the low field (±0.65 T) and the high field regimes. The strong field dependence observed at low fields crosses over to weak field dependence at high fields. Moreover, the slope of the magnetoresistance at high fields decreases as temperature is lowered and eventually changes sign, exhibiting a *negative* magnetoresistance slope, $dR/d|H|$, in a wide field range in the Tesla regime. (As $R(H) > R(0)$ we use the term 'negative slope' rather than 'negative magnetoresistance'). Remarkably, this negative slope is sensitive to the bias current. This is demonstrated in Fig. 4 which shows the magnetoresistance

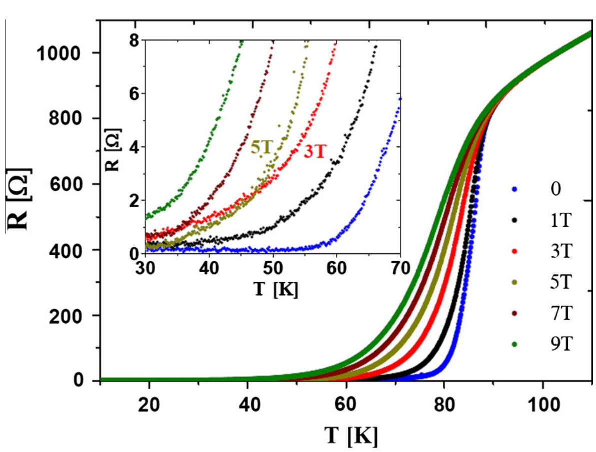

Fig. 2. (Colored in the web version) Temperature dependence of the bridge resistance at different perpendicular magnetic fields, measured with bias current of 2 μA. Inset: Zoom in of the magnetoresistance curves at low temperatures showing crossing of some curves.

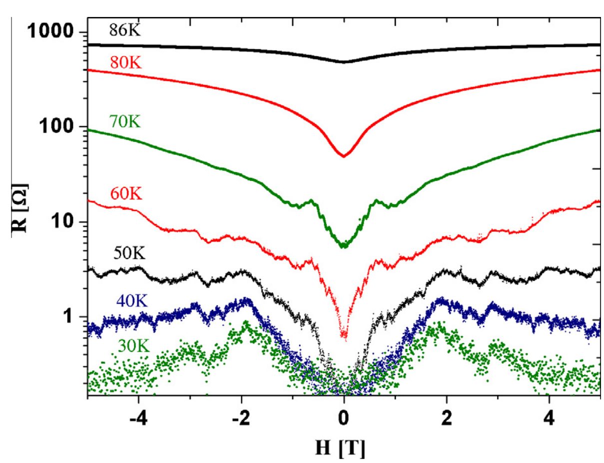

Fig. 3. (Colored in the web version) Semi-log plot of the magnetoresistance of the 500 nm wide wire at the indicated temperatures, measured with bias current of 2 μA. The vertical dashed line points to the onset of the matching-field effect (~0.65 T). The high-fields negative magnetoresistance slope is apparent at low temperatures.

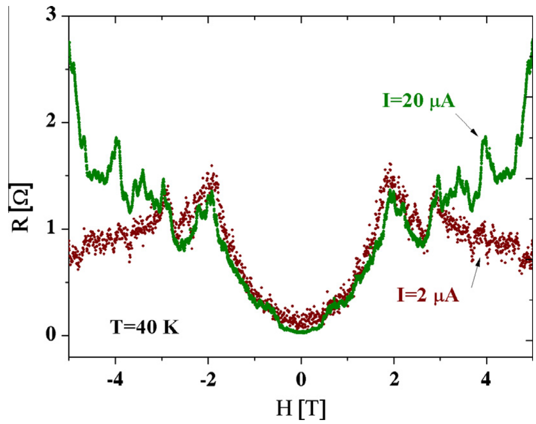

Fig. 4. *R–H* curves at $T = 40$ K for 2 μA (brown) and 20 μA (green) bias current. The negative slope of the magnetoresistance background changes to positive for the higher current. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

at 40 K for bias currents of 2 and 20 μA. Apparently, the slope turns to be positive as the bias current increases. The peculiar behavior of the magnetoresistance described above points to different underlying mechanisms governing the magnetoresistance, as will be discussed below.

Fig. 3 also demonstrates oscillations that are superimposed on the magnetoresistance background. Oscillations in the high field regime with field-periodicity of ~1 T can be identified. The oscillations at low fields are better illustrated in a linear plot of Fig. 5, zooming at the magnetoresistance measured at 70 K between ±0.65 T. A dominant field-periodicity of ~0.12 T can be identified. A closer look at the magnetoresistance data in the low field regime (inset to Fig. 5) reveals a central peak with negative magnetoresistance ($R(0) > R(H)$), starting at zero field and up to $H \approx 500$ Oe. Note that this central peak is symmetric around zero field excluding the possibility that the observed negative magnetoresistance is merely a shift due to residual magnetic field or trapped flux.

Finally, we note that magnetoresistance oscillations and negative magnetoresistance were observed in all the bridges with width between 80 and 500 nm and were discussed in detail in Ref. [6]. The phenomenon which is the focus of this paper, namely the negative slope in the magnetoresistance background, was observed in all the bridges except the narrowest one (80 nm) for a reason to be discussed below. We also note that our preliminary measurements in thicker bridges (>20 nm), in the field and temperature ranges described here, show a monotonically increase of the magnetoresistance background with field; neither a flat region nor a negative slope are found in these thicker bridges. Possible reasons for these observations are discussed below.

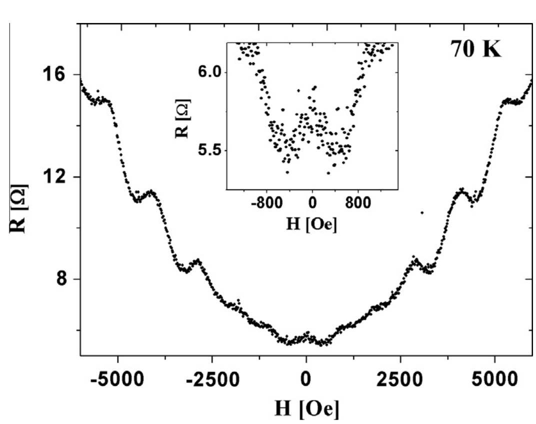

Fig. 5. Magnetoresistance measured at 70 K for fields -0.6 T < H < 0.6 T, showing the oscillations and the background for bias current of 2 μA. Inset: Zoom in on the low-field limit showing the negative magnetoresistance.

4. Discussion

The complex behavior of the background indicates the involvement of several dissipation mechanisms dominating at different temperature and field ranges. We propose below that the dominant mechanisms include phase slips in a distribution of strongly and weakly linked grains dominating at high temperatures (discussed in Section 4.1 below), and quasiparticles tunneling between grains dominating at low temperatures and high fields (Section 4.2).

4.1. Magnetoresistance background at high temperatures

The magnetoresistance depends directly on phase fluctuations rate which increases with increasing magnetic field. Thus, the magnetoresistance background is expected to increase monotonically with the field. To illustrate this behavior we apply Tinkham's expression [7] for the field and temperature dependence of the resistance due to thermally activated phase slippage [8]:

$$R/R_n = \left\{ I_0\left[A(1-t)^{3/2}/2H \right] \right\}^{-2} \tag{1}$$

where I_0 is the modified Bessel function, R_n is the normal resistance, $t \equiv T/T_c$ (in our system $T_c = 89.5$ K), $A \approx 3.5J_{c0}$, $J_{c0}(0)$ is the critical current density at $T = 0$ and $H = 0$. The field H and the parameter A are in units of G, the temperature T in Kelvin and J_{c0} is in units of A/cm^2. In this theory, J_{c0} and R_n are the material parameters which determine the field and temperature dependence of the resistance. Tinkham's prediction for the field dependence of the resistance is illustrated in Fig. 6 for different temperatures. Note that as temperature is lowered the onset of magnetoresistance is shifted to higher fields, and the field at which the magnetoresistance approaches saturations increases.

Attempts to fit Eq. (1) to the data of Fig. 3 – considering our sample as an effective medium characterized by single effective J_{c0} and R_n – were unsuccessful. This is not surprising as our granular sample consists of a distribution of weak links with different J_c. To illustrate the possibility to obtain a background similar to that observed experimentally, we assume that the distribution of weak links in our sample may be represented by only *two* components with different J_{c0} and R_n values. Hereafter we refer to the component with relatively small critical current as 'soft' and that with the higher critical current as 'hard'. The combined effect of the

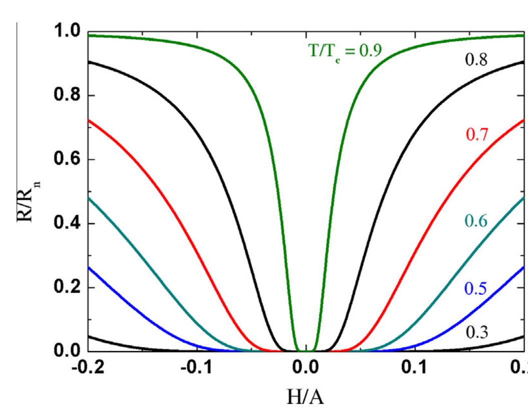

Fig. 6. (Colored in the web version) Dimensionless plot of the magnetoresistance *vs.* field at different temperatures, derived from Eq. (1).

two components is illustrated in Fig. 7 for the 70 K magnetoresistance data. The dashed-blue and the dotted-green lines in the figure (denoted by "soft" and "hard", respectively) are plots of Eq. (1) taking $J_{c0} = 5 \times 10^4$ and 5×10^5 A/cm^2 and $R_n = 18$ and 122 Ω for the soft and hard components, respectively. The dashed-dotted red line is a superposition of the soft and hard components. Apparently, the behavior of the magnetoresistance is controlled by the soft component at low fields and by the hard component at high fields. Note that the dips in the experimental data around ±1 T represent oscillations and are not part of the background. Obviously, the use of only two representative components of soft and hard links is a very crude approximation of the real situation and yet the calculated resistance (the red line in the figure) resembles that of the experimental data, thus demonstrating the applicability of this approximation to our ultrathin sample. Most probably this approximation is not valid for thick samples where a wider and continuous distribution of weak links is expected.

Analysis similar to that shown above for 70 K can be performed for lower temperatures down to 50 K. As demonstrated in Fig. 5, as temperature is lowered the contribution of the hard component to the magnetoresistance becomes significant only at higher and higher fields. As a result, already at 50 K the hard component does not contribute to the magnetoresistance in the field range of the measurement (up to 5 T) and the observed magnetoresistance plateau above ~1.9 T reflects the approach of the soft component to its R_n. Such a plateau is obviously expected also at lower temperatures. Thus, although the Tinkham's phase slips model can explain the data down to 50 K, it cannot explain the negative magnetoresistance slope observed at 40 K and below. It is, therefore, necessary to introduce an additional mechanism in order to explain the magnetoresistance data in this temperature range.

4.2. Magnetoresistance background at low temperatures

To explain a negative dR/d|H| observed in ultra-high fields (tens of Tesla) in $Bi_2Sr_2CaCu_2O_{8+d}$ *crystals* [9], (see also similar results for $(LaSe)_{1.14}(NbSe_2)$ crystals in Ref. [10]), Morozov et al. [9] pointed out that the *c*-axis conductivity in a *d*-wave superconductor is a parallel, two-channel tunneling process between neighboring layers, i.e. tunneling of Cooper pairs and of quasiparticles in gaped and gapless regions, respectively. The conductivity associated with the former decreases with field while that of the later increases [11]. Thus, at high fields the tunneling conductivity of quasiparticles across the CuO_2 layers dominates, resulting in a negative slope in $R(H)$. This model cannot be applied directly to our case as in our bridges the conductivity is in the *a*–*b* plane and, therefore, conductivity through gapless regions is not feasible [12].

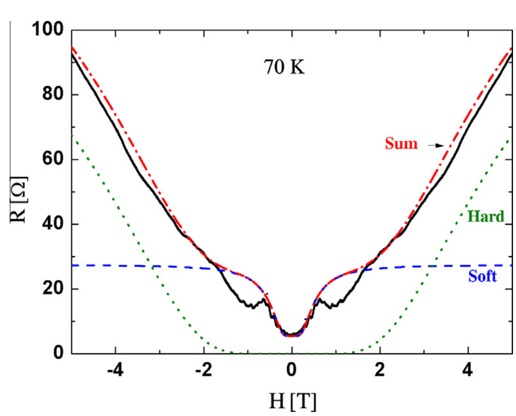

Fig. 7. (Colored in the web version) Magnetoresistance data at $T = 70$ K. The solid blue and green lines are fits to Eq. (1) for the "soft" and "hard" components, respectively. The red line (denoted as "Sum") is the sum of these two. The dips in the experimental data around ±1 T represent oscillations and are not part of the background. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Another model was proposed to explain the negative magnetoresistance slope in the Tesla regime reported for Pb ultrathin films [4], tungsten-based nanowire and ultrathin TiN networks in perpendicular fields [5]. The model [5] emphasizes the significance of the confined geometry in which the magneto-transport properties at high fields are strongly affected by surface superconductivity. At these fields vortices settle at the middle of the wire, and individual Abrikosov vortices merge into a large 'hypervortex' with an extended common core. As a result, this part of the wire becomes resistive, but the resistance is shunted by superconducting channels at the edges. Moreover, formation of hypervortices makes the edge superconducting channels more stable against phase slips and thus gives rise to enhancement of superconductivity. Note, however, that the theory proposed in [5] was developed for homogeneous samples and it is not clear if it is relevant to our granular system.

To explain the negative magnetoresistance slope observed in our YBCO granular bridges we adopt the basic idea of the model proposed in Ref. [9], associating the negative magnetoresistance slope with tunneling of quasiparticles between grains. However, in contrast with the model of Ref. [9] where the negative slope is a result of the interplay between quasiparticles tunneling and pairs tunneling, in our model pair tunneling does not play a role. The mechanism involved are rather phase slips in weak links that saturates at a certain field range and quasiparticles tunneling the rate of which is enhanced by the field. Also, in our case the origin of the quasiparticles is in the normal core of the vortices rather than in the layers as in Ref. [9]. As the field increases, more vortices enter the grains and the number of quasiparticles and the density of states increase, enhancing conductivity of tunneling by quasiparticles [13]. At high temperatures the magnetoresistance increases with the field and the small negative contribution from the quasiparticles tunneling is masked by the strong positive slope. At low temperatures, however, the small contribution of the quasiparticles tunneling can be exposed on the flat, field independent contribution of the phase slips to the background. It is expected that the absence of a flat, field independent region in thick bridges obscures the negative contribution of the quasiparticles.

The effect of the bias current shown in Fig. 4 supports our model. This figure shows that as the bias current is increased from 2 to 20 μA, the negative slope of the magnetoresistance totally disappears. Apparently, the increased current has a similar effect as increasing temperature; both enhance the phase slips rate in the hard component of the weak links. As a result, the effect of tunneling of quasiparticles is less significant and unobservable. The absence of the negative magnetoresistance slope in narrow (80 nm) bridges can be explained in the same way. As the same current range was used to measure the magnetoresistance in all the bridges, the bias current *density* was the largest in the narrowest wire, enhancing the phase slips rate and thus generating a positive magnetoresistance slope which masks the negative contribution from quasiparticles tunneling.

5. Summary and conclusions

The complex magnetoresistance background in granular YBCO bridges is attributed to thermally activated phase slips in weak links and quasiparticles tunneling between grains. At high temperatures the first mechanism is dominant giving rise to an increase in the magnetoresistance due to enhanced phase slips rate with the field. Below a certain temperature the phase slips rate at high fields reaches a saturation, giving rise to a constant magnetoresistance. In this temperature range the contribution of quasiparticles tunneling is revealed, exhibiting a negative magnetoresistance slope due to the increase in the number of quasiparticles with field.

Acknowledgements

Important discussions with Lev Bulaevskii, Valerii Vinokur, Aviad Frydman, Moshe Goldstien and Boris Ya Shapiro are acknowledged. We also thank Irena Shapiro for participating in the analysis of the data and Elran Bruch-El and Omri Sharon for extensive help in wires preparation and measurements. We acknowledge support of the Deutsche Forschungsgemeinschaft through a DIP project. Y.Y. acknowledges a support of the Israel Science Foundation (Grant No. 164/12).

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