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Periodic negative magnetoresistance in granular $YBa_2Cu_3O_{7-\delta}$ nanowires

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Abstract – Magneto-transport measurements in $YBa_2Cu_3O_{7-\delta}$ nanowires, comprising a single layer of a small number of grains, reveal periodic magnetoresistance oscillations in a wide temperature range below the transition. The rich structure of these oscillations manifests a negative magnetoresistance in the initial part of each cycle beginning at zero magnetic field. The salient features of these unique oscillations indicate the presence of π Josephson junctions in the granular wires, arising from the *d*-wave pairing symmetry in $YBa_2Cu_3O_{7-\delta}$.

Superconducting nanowires have become a subject of growing fundamental and applied studies. The applied research is motivated by a desire to apply superconducting nanowires in future electronic circuits and devices. The fundamental studies aim at investigating the behavior of quasi–one-dimensional (1D) superconducting nanowires with diameter comparable to the zero-temperature coherence length, ξ_0. Naturally, most of the fundamental studies have been focused on conventional, low-T_c superconductors due to their relatively large ξ_0. These studies revealed pronounced broadening of the resistive transition associated with thermally activated and quantum phase slips [1]. Measurements in granular nanowires revealed a variety of additional intriguing phenomena such as magnetoresistance oscillations [2,3] negative magnetoresistance [4], anomalous increase in the zero-field resistance near T_c [5], high-field enhancement of the critical current [6], "antiproximity effect" [7], and a magnetic-field–induced re-entry into the superconducting state of current-carrying wires [8].

The small value of ξ_0 in the high-temperature superconductors (HTS) makes the fabrication of 1D wires a rather formidable task. This may explain the relatively small number of studies related to high-T_c nanowires. Nevertheless, few studies of the behavior of HTS wires in less restricted geometries have yielded some fascinating results, so far not fully understood. Jiang *et al.* [9] reported on ultrahigh critical current densities in $YBa_2Cu_3O_{7-\delta}$ (YBCO) narrow bridges with size down to 50 nm. Bonetti *et al.* [10] reported on telegraphic like fluctuations in the resistance of under-doped YBCO wires of width down to 100 nm, at temperatures between the pseudo-gap temperature T^* and the superconducting transition temperature T_c. Mikheenko *et al.* [11] reported on the observation of a transition between the superconducting and resistive states in YBCO wires with widths ranging from 500 down to 50 nm. Xu and Heath [12] succeeded in fabricating YBCO wires with width as small as 10 nm and observed transition broadening due to thermally activated phase slips, with negligible effect of external magnetic field up to 5 T. The electronic properties of HTS *granular* nanowires are expected to be fundamentally different from those of conventional superconductors due to the *d*-wave symmetry of the order parameter in HTS [13,14]. Yet, phenomena associated with the granularity of HTS nanowires have been largely ignored. Misorientation of neighboring grains in such wires can create Josephson junctions with magneto-transport characteristics originating from phase jumps at the grain boundaries. In wide wires, however, the effects of such junctions on transport properties are normally obscured by the presence of large distribution of conducting channels through grain boundaries. Reducing the conduction channels in the restricted geometry of nanowires is expected to expose such effects.

In this paper we report on magnetoresistance measurements in thin (~10 nm) granular YBCO wires with reduced width down to ~80 nm, which is of the order of

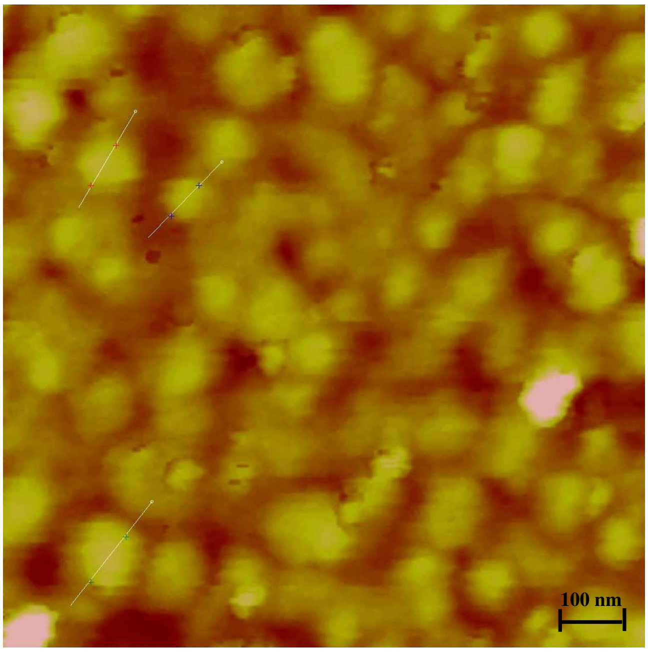

Fig. 1: (Color online) AFM image of $1 \times 1\,\mu m^2$ of the granular YBCO film. Bright colors represent higher areas.

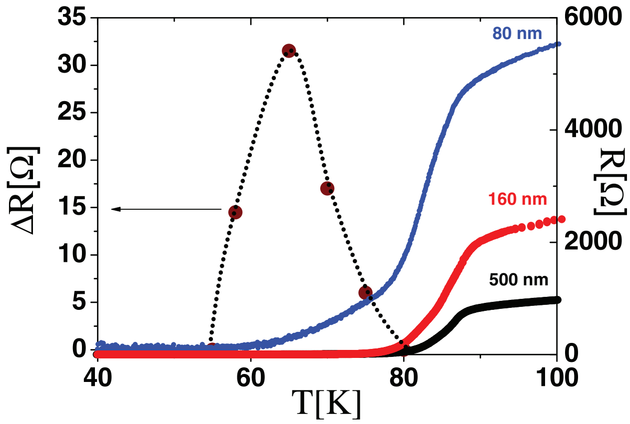

Fig. 2: (Color online) Temperature dependence of the zero-field resistance of 80, 160 and 500 nm YBCO wires (blue, black and red lines, respectively), measured with bias current of $5\,\mu A$ (right ordinate). The brown dots denote the amplitude of the low-field magnetoresistance oscillations, ΔR, for the 80 nm wire (left ordinate). The dotted line is a guide to the eye.

the grain size. Our motivation in preparing such wires was to confine the current path to a channel of width of the order of a grain size, thus minimizing the available conduction channels. These wires revealed unique magnetoresistance oscillations with a rich structure, including a periodic component which exhibits a negative magnetoresistance at the beginning of each cycle, starting at zero magnetic field. Different mechanisms have been proposed to explain the negative magnetoresistance in conventional superconductors (see, *e.g.*, refs. [15–18]). However, these mechanisms explain the negative magnetoresistance *per se*, requiring a different, independent mechanism for the magnetoresistance oscillations. Here, we propose a model that explains *both* the negative magnetoresistance and its periodic behavior found in our experiments. This model invokes zero and π-SQUIDs (superconducting quantum interference devices) that can be created naturally in high-T_c granular superconducting wires.

Pulsed Laser Deposition technique was used to grow 10 nm thick optimally doped c-axis–oriented YBCO films on $10 \times 10\,mm^2$ (100) $SrTiO_3$ wafers. This technique resulted in granular YBCO films with grain sizes between 50 to 100 nm as demonstrated in the Atomic Force Microscopy (AFM) image shown in fig. 1. In most of the grains the c-axis is oriented perpendicular to the film plain, but 3–5% have the c-axis in the plain.

The films were spin-coated with poly(methyl methacrylate) (PMMA) electron-beam resist, and subsequently baked for 90 s on a hotplate at 180 °C. The desired wire pattern, including the pad-contacts for the four-probe measurements, was exposed in the PMMA layer using a CRESTEC CABLE-9000C high-resolution electron-beam lithography system. 700 nm long wires, with width varying between 500 and 80 nm, were prepared. The pads are $\sim 15 \times 15\,\mu m^2$ extension of the wires. After removing the exposed PMMA using methyl isobutyl ketone (MIBK), a mask that defined the wire was transferred to the superconducting film by removing the uncovered parts using a standard argon ion milling process. A Quantum Design Physical Properties Measurements System PPMS MODEL 6000 was exploited to measure the wire resistance in the temperature range between 10 and 300 K and magnetic fields up to 5 T, using DC bias currents between 250 nA and $20\,\mu A$. The magnetic field was applied normal to the film surface (parallel to the crystallographic c-axis).

Standard I-V curves measurements were performed for the 500 and 80 nm wide wires. The critical currents at 10 K, deduced from these curves (not shown), are $\sim 9 \cdot 10^6\,A/cm^2$ and $\sim 2 \cdot 10^6\,A/cm^2$ for these wires, respectively. The value of the critical current for the wider wire is consistent with values reported in the literature for similar films. The difference between the two is a result of the difference in the phase fluctuations rate. Figure 2 shows the temperature dependence of the zero-field resistance of representative YBCO wires with width 80, 160, and 500 nm. The onset of superconductivity is at approximately 88 K for all wires, reflecting the transition to a superconducting state in the grains. As expected for a granular system, the transition width, ΔT, (defined as the temperature range in which the resistance drops from 80% to 20% of its normal value) increases as the wire width narrows down; ΔT is 2, 5, and 14 K for the 500, 160, and 80 nm wires, respectively. The relatively sharp initial transition is followed by a long "tail" characteristic of phase fluctuations in granular materials. In all samples, magnetoresistance oscillations appear in the temperature range where this tail is observed, although with different periods,

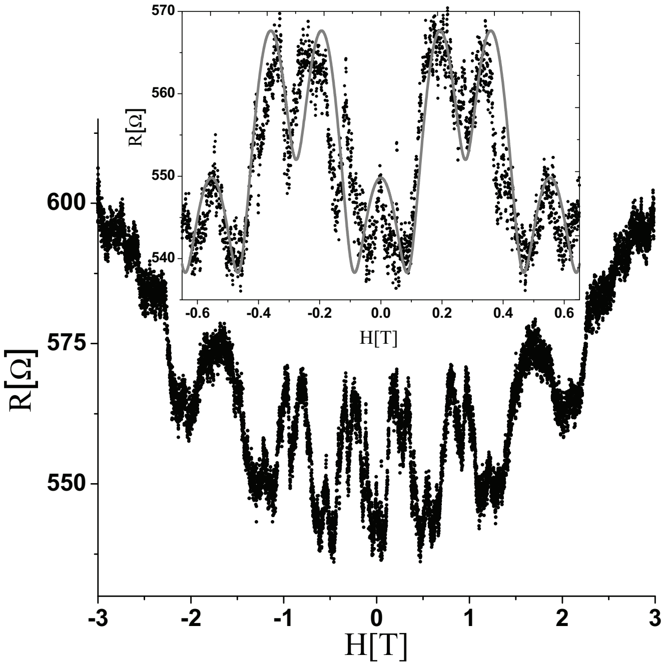

Fig. 3: (Color online) Magnetoresistance oscillations measured at 65 K in 80 nm wide YBCO wire with 5 μA bias current. Inset: Zoom on the first two central periods showing the negative magnetoresistance. Solid line: Calculated magnetoresistance oscillations of a serial connection of zero-zero and zero-π SQUIDs, eq. (1).

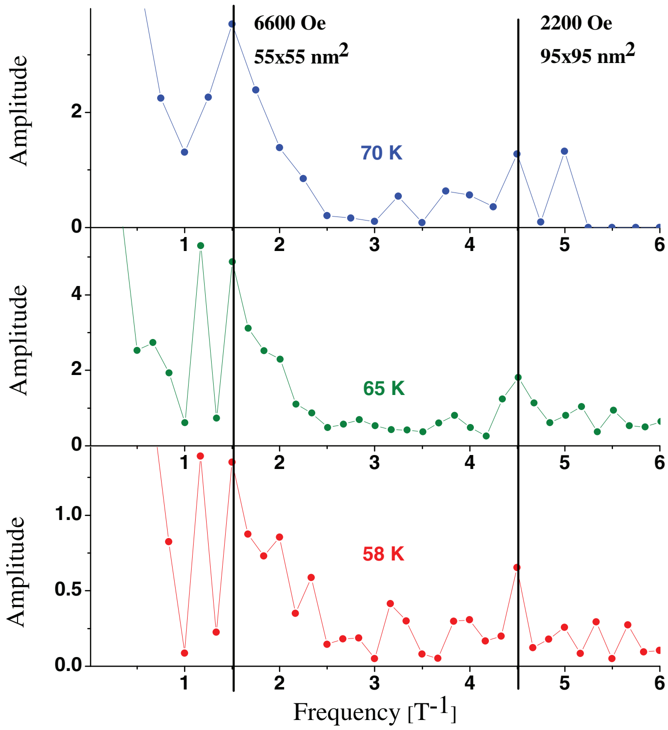

Fig. 4: (Color online) Fast Fourier Transform of the magnetoresistance oscillations measured at the indicated temperatures in 80 nm wide YBCO wire with 5 μA bias current. The vertical lines indicate the two main field periodicities that are temperature independent.

amplitudes and wave forms. In this paper we focus on the narrowest (80 nm) wire which exhibit well resolved oscillations with relatively large amplitude superimposed on relatively slow-varying background.

Figure 3 shows the magnetoresistance oscillations measured at 65 K in a 80 nm wide YBCO wire. We note that the noise that is apparent in the figure appears only in the limited temperature range where the oscillations are observed, reflecting an intrinsic noise, a phenomenon that requires a further study. The rich structure of the oscillations shown in fig. 3 exhibits a central peak with negative magnetoresistance $(R(0) > R(H), \mathrm{d}R/\mathrm{d}H < 0)$, 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. Remarkably, the negative magnetoresistance appears periodically up to $\sim \pm 1.2$ T, namely the periods of the oscillations up to this field exhibits a branch for which the resistance is lower than the zero-field resistance. When the background is subtracted this range is further extended up to approximately 2 T. This periodic feature distinguishes the oscillations observed in our YBCO wire from all the previously reported oscillations in low-T_c wires. Fast Fourier Transform (FFT) analyses of the oscillations obtained at three different temperatures, see fig. 4, yield two dominant, temperature-independent "frequencies", $1/H_{\mathrm{period}}$, associated with field periodicity of approximately 2200 and 6600 Oe [19].

It is tempting to associate the magnetoresistance oscillations with the Little-Parks (LP) effect [20], namely oscillations of T_c due to periodic screening currents in closed loops formed by grains. However, the oscillations in our experiment are observed far below T_c within the resistance tail where $\mathrm{d}R/\mathrm{d}T$ is quite small, see fig. 2. In the LP scenario this will give rise to oscillations amplitude, $\Delta R = (\mathrm{d}R/\mathrm{d}T)\Delta T_c$, being two orders of magnitude smaller than that observed experimentally. Moreover, the LP scenario cannot account for the observed periodic negative magnetoresistance. The occurrence of magnetoresistance oscillations in the temperature range of the resistance tail suggests an alternative explanation, namely that the underlying physics is phase fluctuations in grain boundaries [2,21]. The measured resistance depends directly on the phase fluctuations rate which increases with increasing magnetic field and current. Thus, as the field increases the magnetoresistance is expected to increase monotonically with the field, giving rise to the magnetoresistance background. The oscillations superimposed on this background are generated by screening currents through phase coherent loops of grains [2]. These screening currents are oscillatory with the field due to fluxoid quantization and the requirement for energy minimum [22]. The oscillatory screening currents modulate the rate of phase fluctuations, giving rise to oscillatory magnetoresistance. At very low temperatures the phase fluctuations rate is low and therefore the oscillatory effect of the screening current is negligible. At high temperatures, phase coherence is lost and as a result oscillatory screening currents are absent. Thus, the magnetoresistance oscillations are observed in a limited temperature range dominated by phase fluctuations in phase coherent loops. The effective area, S, of such a loop is associated with a field periodicity, $H_{\mathrm{period}} = \Phi_0/S$, where Φ_0 is the flux quanta. The FFT analysis shown in fig. 4, thus yield effective areas with radius of 55 and 95 nm, consistent with the grain size and the wire width, respectively. Although the above model provides qualitative explanation to the observed oscillations it does not explain the negative magnetoresistance.

As indicated above, negative magnetoresistance, unrelated to the oscillations, was observed at low fields in low-T_c nanowires [4]. Several explanations were suggested for this counterintuitive phenomenon, invoking, *e.g.*, thermally activated phase slips [15], additional channels for magnetoresistance that are blocked by relatively low fields [16], and pair scattering from magnetic impurities due to spin polarization in the presence of the field [17]. A model that can be related to high-T_c superconductivity was proposed by Kivelson and Spivak [18]. This model associates the low-field negative magnetoresistance in granular wires with a random distribution of negative and positive Josephson critical currents. This distribution was attributed to strong fluctuations near the superconducting-insulator transition (SIT) but, as is obvious from fig. 2, our wires are deep in the superconducting regime, far away from the SIT. Yet, we point out that a random distribution of negative and positive Josephson supercurrents can be realized in high-T_c superconductors with d-wave pairing symmetry even far away from the SIT. It has been demonstrated [23] that the d-wave pairing symmetry allows realization of zero as well as π Josephson junctions via grain boundaries intersecting domains of different orientations of the crystal lattice. In our epitaxial grown granular films most of the grains are oriented due to registry with the $SrTiO_3$ substrate, reducing the probability for domains of different crystallographic orientations. Nevertheless, the possibility for the presence of π-junctions in our granular wires still exists. As the grains geometry is not uniform, adjacent facets of neighboring grains may be oriented in different directions, resulting in tunnel junctions with various phase differences across the grain boundaries [24]. For example, Josephson π-junctions can be created in the boundary between two grains in one of which the c-axis is in the plain (few percent of the grains grow along the a direction) or in "step-edge" junction, *i.e.*, the boundary between a grain that "climbs" on a neighboring one. Another possibility is the realization of "geometric" Josephson π-junctions as described in ref. [25]. A scenario for geometric π-junctions is possible in our wires, realizing that most of the grains border the wire edges, and adjacent grains may create narrow constrictions along the wire edges. In this case, a π phase shift can be generated assuming quasi-particles reflected at the wire edge while traveling from one grain to the other through the narrow constriction [26]. This is illustrated in the upper section of fig. 5 which shows the Cooper pairs traveling direction (arrows) through a boundary between grains; the boundary forms a narrow constriction near the wire edge resulting in a dominant contribution from pairs which get reflected at the straight boundary line. The reflected particles suffer a sign change of the pairing potential which leads to a π shift [25]. Thus, the model of Kivelson and Spivak [18] may explain the negative magnetoresistance observed in our wires in the low-field limit. However, this model, as well as the other models listed above, do not account for the *periodic* appearance of the negative magnetoresistance observed in our experiment.

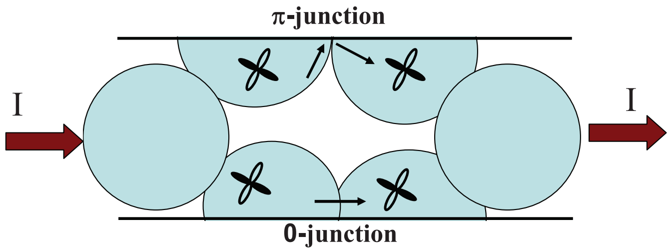

Fig. 5: (Color online) A possible configuration for a π-SQUID in a granular YBCO nanowire. The solid black lines denote the border of a section of the nanowire, including 6 grains. Arrows indicate the travel direction of Cooper pairs through grain boundaries. Cooper pairs traveling through the boundary between the two lower grains do not change their momentum and hence a zero junction is formed. In the boundary between the two upper grains the constriction is narrow resulting in a dominant contribution from pairs which get reflected at the straight boundary line (illustrated by the two small arrows). The reflected particles suffer a sign change of the pairing potential which leads to a π shift, see ref. [25].

In the following we propose a simplified model that attributes *both* the oscillations and the negative magnetoresistance to the *same* physical origin, namely to the presence of π-junctions in our high-T_c superconducting wires. Specifically, we show that the oscillations and the negative magnetoresistance can be produced by a combination of zero and π-SQUIDs. (A π-SQUID comprises zero and π Josephson junctions in parallel, whereas the "conventional" zero SQUID comprises two zero junctions). A possible configuration for a π-SQUID in a granular YBCO nanowire is illustrated in fig. 5. The presence of a π-SQUID is necessary in order to produce the periodic negative magnetoresistance. The FFT of the oscillations (fig. 4) which exhibits two main field periods ($\sim$2200 and $\sim$6600 Oe) suggests that two SQUIDs with areas differing by a factor of $\sim$3 play a role. The geometrical constraints of the wires (ratio of the wire width to the grain size of order 1) force these SQUIDs to be connected in series. The voltage across this serial connection is given by the sum of the voltage drops across the zero and the π-SQUIDs, namely [22,26,27].

$$V = (R_1/2)\sqrt{I^2 - \left(2I_{c1}\cos\frac{\pi SH}{\Phi_0}\right)^2} + (R_2/2)\sqrt{I^2 - \left(2I_{c2}\sin\frac{\pi(3S)H}{\Phi_0}\right)^2}, \qquad (1)$$

where I is the measuring current, H the applied field, S the effective area of the zero-zero SQUID, R_1 and R_2 are the single-junction resistances, and I_{c1} ,I_{c2} the critical currents of the Josephson junctions in the zero and π-SQUIDs, respectively, (assuming that the two junction in each SQUID have the same resistance and critical current, and ignoring the self-inductance of the loops). The solid line in the inset to fig. 3 is a plot of eq. (1) taking $I_{c1}/I = I_{c2}/I = 0.48$, $R_1 = R_2 = 60\,\Omega$ (see footnote [1]), and $S = 3300\,\text{nm}^2$ corresponding to a length scale of $\sim$55 nm, typical of a grain size; $3S$ corresponds to a length scale of 95 nm, close to the wire width. Evidently, the calculated curve shown in the inset to fig. 3 captures the experimentally observed negative magnetoresistance and the oscillations. Obviously, this model does not attempt to resolve all the details of the oscillations but only to capture their salient features that indicate the presence of π-junctions in the system. Our simplified model also explains the limited range for which the oscillations are observed. The voltage swing generated by each SQUID is determined by the ratio $I/2I_c$ and it is maximum for $I = 2I_c$. At high temperatures for which $I \gg 2I_c$ the oscillations amplitude drops; at low temperature I becomes smaller than $2I_c$ and consequently no voltage is generated. It should be noted that additional weakly linked chains of grains which may be connected in series to the two SQUIDs may contribute to the background and not to the periodic oscillatory behavior.

In summary, transport measurements in YBCO nanowires revealed periodic magnetoresistance oscillations displaying a unique behavior, namely negative magnetoresistance in the initial part of each cycle beginning at zero field. We attributed these oscillations to the granular nature of the nanowires and to the d-wave pairing symmetry in the grains. Reducing the wire width down to the order of the grain size filters out many oscillatory components of different frequencies, revealing the important role played by zero and π Josephson junctions in producing the magnetoresistance behavior. We further showed that the salient features of the oscillations obtained for the narrow wire can be modeled as a serial combination of zero and π-SQUIDs. This model is unique in the sense that it suggests a single mechanism for both the oscillations and the negative magnetoresistance, the latter being a consequence of the π-junctions that can be found in high-T_c granular wires.

* * *

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[1] $R_1 = R_2 = 60\,\Omega$ is consistent with the geometry of the wire. If the system may be crudely approximated as a 700 nm long 1D wire, a loop of diameter of $\sim$55 nm in this wire is expected to contribute $\sim$8% of the total resistance of $\sim$560 Ω, *i.e.*, $\sim 45\,\Omega$.

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