Quantum fluctuations in ultranarrow superconducting aluminum nanowires

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Progressive reduction of the effective diameter of a nanowire is applied to trace evolution of the shape of the superconducting transition $R(T)$ in quasi-one-dimensional aluminum structures. In nanowires with effective diameter $\leqslant 15$ nm the $R(T)$ dependences are much wider than predicted by the model of thermally activated phase slips. The effect can be explained by quantum fluctuations of the order parameter. Negative magneto-resistance is observed in the thinnest samples. Experimental results are in reasonable agreement with existing theoretical models. The effect should have a universal validity, indicating a breakdown of the zero-resistance state in a superconductor below a certain scale.

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I. INTRODUCTION

The ability to carry a dissipationless electric current is a fundamental attribute of a superconductor. One might naively expect that this property is preserved at reduced dimensions. Unfortunately, inevitable fluctuations result in momentary suppression of superconductivity, leading to energy dissipation. Being integrated in time, this effect manifests itself as finite resistance. Of particular interest are quasi-one-dimensional (quasi-1D) systems, with effective diameter $\sigma^{1/2}$ smaller than the superconducting coherence length ξ, where there is only one parallel channel of supercurrent. In such objects, due to fluctuations of the order parameter, the effective resistance is never strictly equal to zero. However, a measurable resistance at temperatures well below the critical temperature T_c can be observed only in samples with rather small diameters. Early experiments[1] confirmed that in narrow superconducting channels the shape of the $R(T)$ transition can be explained by thermal fluctuations.[2] Recent experimental[3–8] and theoretical[9–19] discoveries claim the existence of an additional (nonthermal) mechanism of finite resistance in 1D systems. The issue is of vital importance for the development of superconducting nanoelectronic elements designed to carry a dissipationless electric current.

The finite resistance of a 1D channel can be understood that for sufficiently long systems of length $L \gg \xi$ there is always the finite probability of a fluctuation to drive instantly a fraction of the wire of volume $\xi\sigma$ into a normal state. The energy required for this process is the corresponding superconducting condensation energy $\Delta F \sim B_c^2 \xi\sigma$, where B_c is the critical magnetic field. If thermal effects solely contribute to the fluctuations, it has been shown[2] that the effective dc voltage $V(T,I)$ is proportional to the probability of these events $\sim \exp(-\Delta F / k_B T)$, where k_B is the Boltzmann constant:

$$V(T,I) = \Omega(T,I,L)\exp\left[-\frac{\Delta F}{k_B T} - \left(\frac{2}{3}\right)^{1/2}\frac{I^2}{3\pi I_0 I_c}\right]\sinh\left(\frac{I}{2I_0}\right),$$

(1)

where I_c is the temperature-dependent critical current, and $I_0 = k_B T / \phi_0$ with ϕ_0 being the superconducting flux quantum. The current dependence is determined by the $\sinh(I/2I_0)$ term and the term under the exponent, responsible for the

effective reduction of the potential barrier ΔF by the finite current I. An exact form of the prefactor $\Omega(T,I,L)$ has been proposed.[2] Though the function $\Omega(T,I,L)$ also contains temperature- and current-dependent terms, it contributes negligibly compared to the strong exponential dependence. Formally the above process can be described as the thermally activated transition of a superconducting system from a local potential minimum to a neighboring one separated by $\pm 2\pi$ in phase space: so-called, thermally activated phase slip (TAPS). One can make a formal analogy with a "classical jump" of a particle over the barrier ΔF stimulated by the thermal energy $k_B T$.

An alternative mechanism has been proposed associated with tunneling through the energy barrier ΔF in phase space.[3] Such tunneling to a state of lower energy, also called *quantum phase slip* (QPS), should provide an additional (to TAPS) channel of energy dissipation in a current-carrying system. For conventional type-I superconductors estimations show that measurable deviations from the TAPS mechanism are expected in samples with effective diameter $\sigma^{1/2} \sim 10$ nm. There have been a few experiments claiming observation of the QPS phenomenon in various superconducting materials: In, Pb, and Pb-In,[3] Pb, Sn, and Pb-Bi,[4] MoGe,[5] Sn and Zn,[6] and Al.[7,8] Various models consider nonthermal mechanisms of the finite resisitivity of a 1D superconductor.[9–19] In spite of the intensive research in the field, the matter is far from being settled. From an experimental point of view, to some extent, the inhomogeneity of a nanowire might lead to a broad superconducting transition $R(T)$ which can be erroneously associated with the QPS mechanism.[20] Another source of misinterpretation might come from insufficient filtering from environmental rf noise. The resulting parasitic overheating can shift the superconducting transition to lower temperatures, distorting the shape of the $R(T)$ dependence.

Our motivation was to perform experiments eliminating the uncertainty related to the uniqueness of each particular nanostructure. We were able to trace the crossover from the TAPS mechanism to QPS in the *same sample*, successively reducing its diameter between measurements, thereby evidencing for a solely size-dependent origin of the phenomenon. The experiments performed on multiple sets of aluminum nanowires reduced by ion sputtering down to sub-
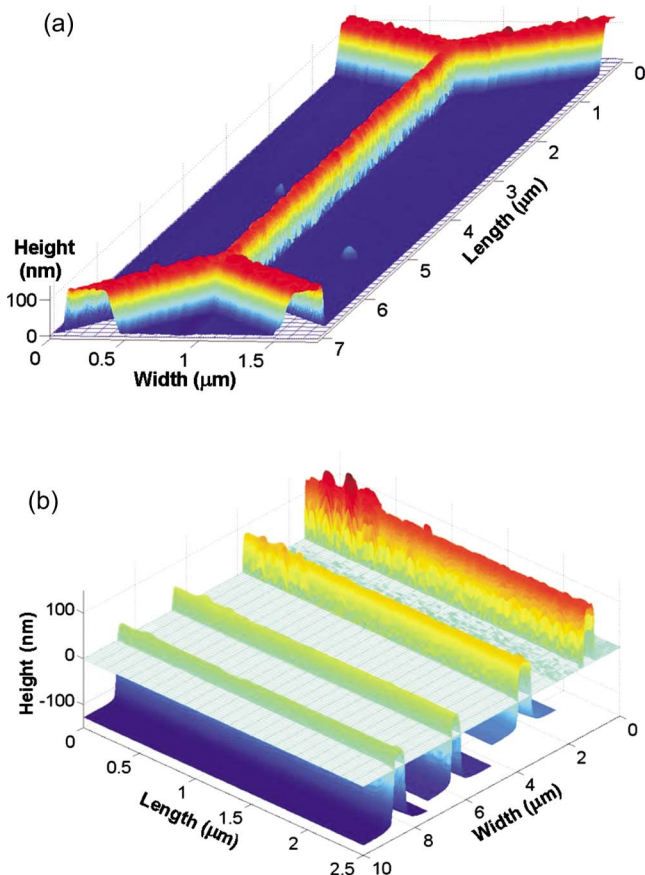
(a)

(b)

FIG. 1. (Color online) (a) Atomic force microscope (AFM) image of a typical 5-μm-long aluminum nanowire just after lift-off. (b) AFM images showing evolution of the shape of the same nanowire after several sessions of ion beam sputtering. Bright color above the horizontal plane (initial level of the substrate) corresponds to metal, dark color below to sputtered Si substrate. Note the reduction of the initial surface roughness of the nanowire.

10-nm scales showed quantitatively similar behavior. In all structures with effective diameter $\sigma^{1/2} < 15$ nm the $R(T)$ dependences deviate from the predictions of the TAPS model,[2] while the QPS scenario[11] provides a reasonable fit to the data.

II. EXPERIMENT

In our earlier work[21] we demonstrated that low-energy Ar$^+$ ion sputtering progressively and nondestructively reduces nanostructure dimensions. The penetration depth of Ar$^+$ ions into an Al matrix at acceleration voltages of ~500 eV is about 1.5 nm and is comparable to the thickness of naturally formed oxide. The accuracy of the effective diameter $\sigma^{1/2}$ determination from the normal-state resistance, scanning electron microscopy (SEM), and scanning probe microscopy (SPM) measurements is about ± 2 nm. Only those samples which showed no obvious geometrical imperfections were used for further experiments. While reducing the nanowire diameter, the low-energy ion sputtering also provides "polishing," removing surface roughness inevitable after a lift-off process[7] (Fig. 1). In this work the method[21,25]
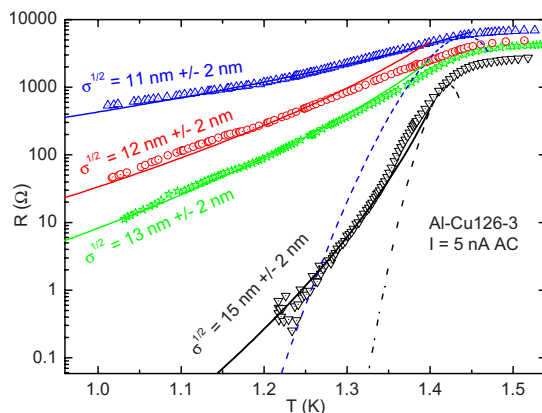

FIG. 2. (Color online) Resistance vs temperature for the thinnest samples obtained by progressive reduction of the diameter of the same aluminum nanowire Al-Cu126-3 with length $L=10$ μm. The Langer-Ambegaokar-McCumber-Halperin (LAMH) model fitting is shown with dashed lines for 11 and 15 nm samples with the best-fit mean free path $\ell=3$ and 10 nm, correspondingly, $T_c=1.46$ K and critical magnetic field $B_c(0)=10$ mT. Fitting using a simplified short-wire model (Ref. 11) [Eqs. (2) and (3)] is shown with solid lines. For 11-, 12-, 13-, and 15-nm wires the fitting parameters are $T_c=1.5$ K; $A=0.15$, mean free path $\ell=5.4$, 5.8, 7.3, 7.5 nm; and the normal-state resistance $R_N=7200$, 5300, 4200, and 2700 kΩ.

was applied to lithographically fabricated 99.999% pure Al nanowires with initial cross section of about 100 nm $\times$ 100 nm. During measurements the structures were immersed into a directly pumped ^{4}He bath with base temperature of about 0.95 K. Contrary to Refs. 5, 6, and 8, we used a four-probe configuration, eliminating the necessity to consider the contribution of the electrodes. We obtained exact quantitative agreement between data measured with dc and ac currents using extensive multistage rf filtering. The results are reproducible and do not suffer from hardware-related artifacts.

III. RESULTS AND DISCUSSION

After a sequence of sputterings [alternated with $R(T)$ measurements] the wire cross section σ is reduced down to ≤ 20 nm, where deviations from TAPS behavior become obvious (Fig. 2). Similar results were obtained on several sets of aluminum nanowires with length L equal to 1, 5, and 10 μm. For larger diameters (e.g., Fig. 3, inset, 55-nm wire) the shape of the $R(T)$ dependence can be qualitatively described by the TAPS mechanism. Notice that a size-dependent variation of the critical temperature of a nanowire,[22] especially pronounced in aluminum,[7] results in a broadening of the $R(T)$ transition and significantly reduces the straightforward applicability of the TAPS model.[20] Confidence in assigning our results to the manifestation of a different (nonthermal) mechanism comes from the experimental observation that in aluminum nanowires (films) the critical temperature increases with a decrease of the wire diameter (film thickness).[22,7] It means that the broadening of the $R(T)$ dependences below the bulk T_c value ~1.2 K can-
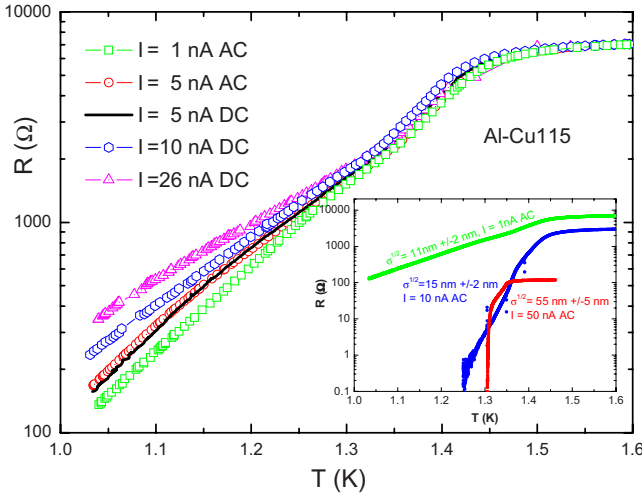
FIG. 3. (Color online) Resistance vs temperature for a $\sigma^{1/2}=11$ nm ± 2 nm sample of aluminum nanowire Al-Cu115 with length $L=10$ μm measured at various dc and ac currents. Inset: resistance vs temperature for three samples obtained by progressive reduction of the diameter of the same nanowire.

not be explained by a geometrical imperfection (e.g., constriction) of an aluminium nanowire. Below a certain limit $\sigma^{1/2} \leq 20$ nm fits by the TAPS model fail to provide any reasonable quantitative agreement with experiment even assuming the existence of unrealistically narrow constrictions not observed by SPM (Fig. 2).

Though several theoretical approaches[9–19] were proposed to describe the non-TAPS mechanism of finite resistance in 1D superconducting channels, not all models are suitable for direct comparison with experiment, lacking a clear expression for the resistance (voltage) as a function of temperature. Where possible, we tried to compare our data with the models and found the best agreement with renormalization theory.[11] The full version of the model for systems of an arbitrary length L is rather sophisticated, requiring knowledge of parameters not easily deductible from experiment. However, if the wire is short enough that only one phase slip event can happen at a time, one can neglect the effects of the interaction between the phase slips. In this limit the simplified expression for the rate of QPS activation is

$$\Gamma_{QPS} = \frac{S_{QPS}L}{\tau_0 \xi} \exp(-S_{QPS}), \qquad (2)$$

where the action $S_{QPS}=A(R_Q/\xi)/(R_N/L)$, with A being a numerical constant, $R_Q=h/(4e^2)=6.47$ kΩ, and $\tau_0 \sim h/\Delta$ is the characteristic response time of a superconducting system which roughly determines the duration of each QPS.[11] The effective (time-averaged) voltage V_{eff} due to the fluctuations can be found using the Josephson relation.[2] Finally, for the effective resistance of a quasi-1D superconducting nanowire one gets

$$R_{QPS}(T) \equiv V_{eff}/I = \frac{h\Gamma_{QPS}}{2eI}. \qquad (3)$$

Contrary to TAPS,[2] the QPS contribution has a rather weak temperature dependence far from the critical temperature and should produce a finite resistance even at $T \rightarrow 0$ for sufficiently narrow wires.[11] One can achieve reasonable agreement between the model [Eqs. (2) and (3)] and the experiment (Fig. 2). There are four fitting parameters: critical temperature T_c, normal-state resistance R_N, mean free path ℓ [to recalculate the dirty-limit coherence length ξ $=0.85(\xi_0\ell)^{1/2}$], and the numerical parameter A of the order of unit.[11] The critical temperature and the normal-state resistance can be trivially deduced from experimental $R(T)$ dependences. Roughly the mean free path ℓ can be estimated from the normal-state resistivity ρ, as the product $\rho\ell=5\times 10^{-16}$ Ω m^2 is a well-tabulated value for dirty-limit aluminum. In our ultranarrow nanowires the cross section is known from SPM measurements with ± 2 nm accuracy. Hence, there is some freedom in the selection of the mean free path. As a rule of thumb for all our nanowires with effective diameter $\sigma^{1/2} \leq 20$ nm the best-fitted mean free path (at low temperatures) was found to be roughly equal to one-half of the diameter (Fig. 2, caption). The observation is quite reasonable, taking into consideration that at these scales and temperatures electron scattering is mainly determined by the sample's physical boundaries. For all aluminum nanowires the best-fitted value for the parameter A was found to be equal to 0.15. As the value of A cannot be calculated within the model[11] with the required accuracy, we believe that the correspondence between the simplified "short-wire" model and the experiment can be considered as good.

No Coulomb blockade has been observed on $V(I)$ characteristics, indicating good homogeneity of the wires (absence of tunnel barriers). Below the critical temperature $V(I)$ dependences show $\sim \sinh(I/I_0)$ behavior similar to both TAPS [Eq. (1)] and renormalization[11] models. At sufficiently small currents $I \ll 2I_0 \sim 20$ nA one might expect a linear response, while at much higher currents trivial overheating is observed. However, there exist an intermediate regime between linear and strongly nonlinear regimes: the top part of the $R(T)$ transition is not shifted, while the slope of the bottom part decreases with an increase of the measuring current (Fig. 3). We associate this peculiarity with the reduction of the potential barrier ΔF by a finite current I [Eq. (1)], which is qualitatively similar for thermal[2] and quantum[11] mechanisms.

At certain limits the renormalization model[11] predicts the same functional dependence of the effective resistance on temperature and current. In the high-temperature limit $T \gg \phi_0 I/k_B$, $R \sim T^{2\gamma-2}$, and in the high-current limit $I \gg I_0$, $R \sim I^{2\gamma-2}$. The dimensionless conductance $\gamma=R_Q/R_{qp}$ is related to the effective "quasiparticle" resistance R_{qp}, being associated with dissipation provided by the quasiparticle channel. R_{qp} can be considered as a fitting parameter and should be of the order of the normal-state resistance R_N. In the metallic phase, when quantum fluctuations dominate the behavior of the system, the residual resistance at $T \rightarrow 0$ should correspond to R_{qp}.[11] The results of fitting of our $R(T)$ and $V(I)$ dependences to the above relations are presented in Fig. 4. One may obtain satisfactory agreement with experimental data, allowing the quasiparticle resistance R_{qp} to be
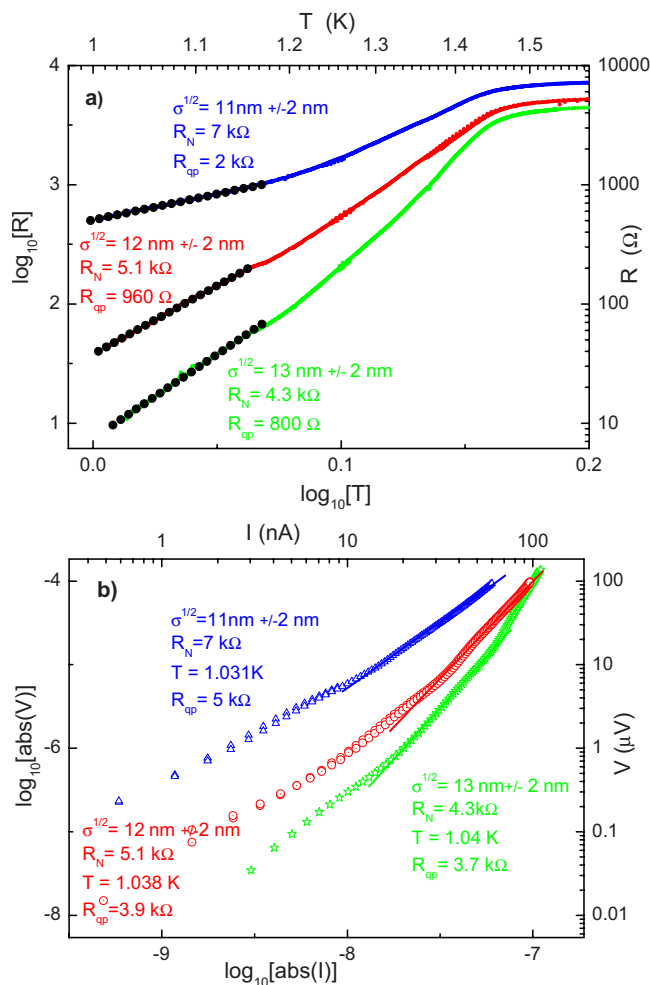
FIG. 4. (Color online) (a) Resistance vs temperature for three samples of the same wire as in Fig. 2 with progressively reduced cross sections. Solid dots are fittings to power dependence $R \sim T^{2\gamma-2}$. (b) $V(I)$ dependences of the same samples taken at close temperatures stabilized with accuracy ± 0.1 mK. Solid lines correspond to proportionality $R(T) \equiv V(T)/I \sim I^{2\gamma-2}$. In both figures the only fitting parameter is the quasiparticle resistance R_{qp}.

smaller than the normal-state resistance R_N of the nanowire. For each sample, the deviation of the fitting parameter R_{qp} from R_N is less pronounced for $V(I)$ characteristics. Probably, this discrepancy is the result of the experimental difficulty to satisfy the applicability condition $T \gg \phi_0 I/k_B$, while the temperature should be noticeably below the critical temperature T_c, where the whole concept of phase slippage is valid.

Application of a magnetic field perpendicular to the plane of the structures reveals a rather unusual effect. At sufficiently small temperatures a pronounced negative magnetoresistence (nMR) is observed (Fig. 5, inset), resulting in a "sharpening" of the $R(T)$ transitions in magnetic fields $\leqslant 25$ mT (Fig. 5). nMR is observed only in the thinnest samples $\sigma^{1/2} \leqslant 20$ nm and only at temperatures well below the onset of superconductivity. A similar effect has been reported for ultrathin 1D lead strips.[4] Aluminum is known to be immune to the creation of localized magnetic moments provided by the majority of magnetic elements. Our samples
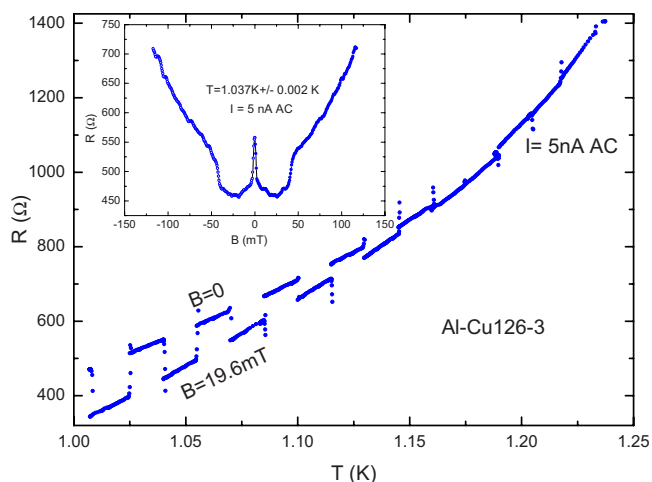

FIG. 5. (Color online) Slowly recorded ($\sim$1 h) resistance vs temperature dependence for the 11-nm sample from Fig. 2. While sweeping the temperature, a few times the perpendicular magnetic field $B = 19.6$ mT was switched on and off. The top branch corresponds to zero field, while the lower one to field "on." Inset: resistance vs perpendicular magnetic field for the same sample measured at constant temperature and small ac current.

were *e*-beam evaporated from a 99.999% pure aluminum target in an UHV chamber where magnetic materials have never been processed. Hence, the residual concentration of magnetic ions should be negligible. Even if we were to assume their presence, magnetic fields where nMR is observed are too small to polarize them. Additionally, we do not see an enhancement of T_c related to the onset of superconductivity, which would be the consequence of suppression of the Kondo mechanism by a magnetic field. As the presence of localized magnetic moments in our samples is not obvious, a recently developed model of nMR (Ref. 23) is not applicable. nMR has been predicted in disordered superconducting wires.[19] However, the main result of that model is an exponentially small addition to the TAPS effective resistance. This conclusion contradicts our main observation (Fig. 2) and, hence, the applicability of the whole model[19] to our experiments is questionable. For the moment we do not have a solid explanation for the nMR effect. The phenomenon might be related to an interplay between two field-dependent contributions: ΔF barrier reduction and suppression of the superconducting energy gap Δ. The first process provides an expected increase of the observed resistance, while the second one leads to a reduced quasiparticle resistance R_{qp} due to thermal activation of extra quasiparticles resulting in lower effective resistance $R(T)$. There might be a region of magnetic fields where the second mechanism "wins," reducing the influence of quantum fluctuations leading to nMR.[11] Another explanation of the nMR might be related to the formation of a charge-imbalance region accompanying each phase-slip event.[24] This nonequilibrium region provides dissipation outside the core of a phase slip. The corresponding ohmic contribution can be effectively suppressed by the magnetic field, resulting in nMR. However, so far the validity of the charge-imbalance concept was only demonstrated at temperatures sufficiently close to T_c and its applicability to QPS

at temperatures well below the critical one is not obvious. A quantitative comparison with experiments requires further elaboration of the theory.

IV. CONCLUSIONS

The homogeneity of the wires is the central point in an interpretation of data related to phase-slip mechanisms: thermal or quantum. The existence of trivial structural or geometrical imperfections as constrictions, boundaries, and contact regions might broaden the $R(T)$ dependences[20] and can be erroneously interpreted as a "new physics." The specially developed method of ion beam sputtering[21,25] to a large extent allowed us to study the evolution of the size phenomenon, eliminating artifacts related to the uniqueness of samples fabricated in independent processing runs. The ion beam treatment polishes the surface of the samples, removing the inevitable roughness just after fabrication (Fig. 1). If there were no detectable geometrical imperfections in the original (thick) wires, they cannot be introduced while reducing the diameter by low-energy ion sputtering. Formally, it cannot be excluded that in the original structures there were "hidden," undetectable structural defects (e.g., highly resistive grain boundaries) which did not contribute to conductivity, being shunted by the bulk. When the diameter of the wire is reduced below a certain scale, this type of imperfections might determine the behavior of the quasi-1D system. However, the absence of Coulomb blockade and/or gap structure on I-V characteristics rules out the presence of the highly resistive tunnel barriers.

The extensive SPM and SEM analysis indicates that after several sessions of ion beam sputtering the surface of our nanowire is flat with few nm accuracy.[25] This is quite small in absolute terms, particularly keeping in mind that the lattice constant in aluminum is $\sim$0.4 nm. However, approaching the 10-nm scale, a few nm roughness means a lot in relative terms. Hence, we cannot exclude the possibility that the specified average effective diameter of our nanowires is not very representative: the phase slippage might happen in the weakest (thinnest?) parts. To eliminate completely this uncertainty one should study infinitely long atomically flat structures, which are quite problematic to fabricate. Nevertheless, the main conclusion of the present paper is still valid even assuming the presence of inevitable imperfection of the samples. At all realistic parameters characterizing the geometry of the possible constrictions, the thermal activation scenario[2] fails to explain the broad $R(T)$ dependences observed in sub-15-nm nanowires, while the QPS mechanism[11] provides good agreement with the experiment (Fig. 2).

In conclusion, we have traced the evolution of the shape of the superconducting transition $R(T)$ in quasi-one-dimensional aluminum nanowires with progressive reduction of their cross sections. For relatively thick samples $\sigma^{1/2} \geqslant 20$ nm the shape of the transitions can be qualitatively explained in terms of wire inhomogeneity and the model of thermally activated phase slips, while in nanowires with effective diameter $\leqslant 15$ nm the $R(T)$ dependences are much wider and no reasonable set of fitting parameters can account for the TAPS mechanism. The phenomenon is associated with manifestation of quantum fluctuations of the order parameter. The results are in good agreement with renormalization theory.[11] The effect of quantum fluctuations should have a universal validity, indicating a breakdown of the zero-resistance state in quasi-one-dimensional superconductors, setting fundamental limitations on miniaturization of nanoelectronic components designed to carry a dissipationless supercurrent.

ACKNOWLEDGMENTS

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