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Suppression of superconductivity in thin Nb nanowires fabricated in the vortex cores of superfluid helium

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ABSTRACT

Nanowires of niobium, platinum and indium–lead $In_{88}Pb_{12}$ alloy with diameters of 4.2, 3.6 and 8 nm, respectively, were grown in quantized vortices of superfluid helium, and the dependences of their resistance on temperature have been studied. Through a detailed comparison of these dependences we present evidence that superconducting niobium wires allow a high rate of quantum phase slip. This phase slippage leads to a phase transition to an insulating state at $T \to 0$.

1. Introduction

Quasi-one-dimensional metals, namely nanowires, are one of the most interesting objects in nanoscale science. Naturally, the size effects are mainly the fingerprints of quantum behavior. However even such a promising and unusual chemical property as high catalytic activity of noble metals is also observed only for diameters of particles in a range of 3–5 nm [1–3], the same scale can be of relevance for nanowires, which can also turn to be catalytic if made sufficiently thin. From the point of view of physics the most attractive properties of nanowires are the size effects in their electrical conductivity and first of all in superconductivity [4–9]. If the wire diameter is made smaller and smaller the critical temperature of the wire approaches zero [10]. However, superconductivity may disappear well before the critical temperature, T_c, becomes zero. In particular, a quantum Kosterlitz–Thoulesse transition, involving a quantum depairing of phase slips and anti-phase-slips, has been predicted to occur in superconducting wires with diameters of just a few nanometers [11]. Such transition should induce a dielectric behavior in a nanowire made of a superconducting metal or alloy. The quantum phase slips (QPS), which is the main reason for the occurrence of the insulating state in nominally superconducting wires, has been discussed in numerous publications [5,6,11,12]. Typically it is accepted to distinguish two types of them: thermally activated phase slips (TAPS) [4] and quantum phase slips [5,7–9,13,14]. The QPS can make a thin wire acts like a weak insulator, even if it is made of a superconducting material [5,6]. It has been suggested that nanowires which support quantum phase slips can be used to make novel logical quantum devices. A proof-of-principle qubit based on a nanowire has been demonstrated recently [14].

The main focus of the present work is the experimental study of thin superconducting nanowires aimed to revealing the quantum size effects in their electrical conductivity. Here we present an observation of an insulating behavior in pure-metal elemental nanowires (Nb), having a surface which is clean and not oxidized. We explain the observation in terms of proliferation of quantum phase slips [11]. It is worthy to remind that other factors, besides QPS, can act as strong suppressants of the superconducting order in ultrathin nanowires. It is well known, since the pioneering works of Abrikosov and Gor'kov [15], that magnetic impurities, if their concentration increases, first reduce the critical temperature, then make superconductivity gapless and finally completely suppresses the critical temperature to zero. In other words, if the concentration of uncompensated spins in the superconductor is sufficient, they can eliminate completely all the superconducting effects including the Meissner effect. Since the surface-to-volume

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ratio becomes especially large in nanowires having diameters of just a few nanometers, contamination of the surface with unpaired spins, of any origin, can have a strong impact on superconductivity. In particular, it was suggested [16] that the nanowire surface, if subjected to oxidation, can develop unpaired spins due to disorder. If unpaired spins are present at the boundary between the metal and the oxide, they can act, effectively, as magnetic impurities on the surface of the nanowire. Of course no actual impurity is present in this case [15], simply the boundary between the metal and its oxide is typically so disordered that unpaired and single-occupied localized electronic states can occur. These "free radicals" or "dangling bonds" potentially may have spins and so they may be qualified as magnetic depairing agents. In order to rule out the surface spin effect with complete certainty one has to perform experiments with nanowires prepared in extremely clean conditions. Also, nanowires should not be exposed to air, even briefly. The method of nanowires production and measurements which we used here conforms to these requirements.

2. Experimental

The difficulties in the experimental study of size effects in superconductivity suppression are associated as well with the fact that these effects should appear for wires with very small diameter, and the production of such a superconductor wire, possessing desirably regular structure, perfect shape and containing no impurities represents rather sophisticated problem. The method of nanowire formation in the cores of quantum vortices in superfluid helium helps to eliminate the concerns listed above.

Our method of nanowires production, using quantized vortices of superfluid helium as a template, allowed us to grow nanowires from virtually every metal and alloy [17]. The quality of the nanowires is quite reasonable, as they are formed by fusion in the quantized vortex core of nanoclusters being molten at the moment of their nucleation by the atom condensation [18]. The diameter of the nanowires formed by this technique is determined mainly by the thermal characteristics of the metal [17] and it ranges from about 7 to 8 nm for fusible metals to as small as 3–4 nm for refractory metals [17,19,20]. It is therefore can be expected that, at least for refractory metals, the diameter of the nanowires would be sufficiently small for certain size effects to become pronounced. As we will discuss in detail below, this expectation is true and our ultrathin Nb nanowires exhibit, indeed, a dielectric behavior, which we associated with proliferation of quantum phase slips [11].

When growing in superfluid helium the nanowires prefer to pin to various protuberances present in the zone of the growth. Indeed, the quantized vortices in superfluid helium tend to connect to sharp pins, in particular to the specifically installed pins ("electrodes") connected to the measurement leads. As a result, the bundles of nanowires were grown between the tips of the electrodes intentionally placed vertically with few millimeters distance between them. Finally the nanowires closed the electrical circuit acting as a bridge connecting the tips of the electrodes. Thus, it was possible to measure the resistance of the nanowire bundles directly in the cryostat, without exposing the nanowires to ambient air. Consequently, both the process of the wires growth and the process of their electrical transport measurements were perform in uniquely clean and inert conditions of liquid helium-4. It is well known that no contaminants and chemical reactions can be present in liquid helium. Therefore the surface of the wires was perfectly clean. As it has been shown in [19], the resistance of a bundle quite reasonably reflects the resistivity of single wire.

Our experimental technique was described elsewhere [17,19]. Atoms and small clusters of the metal are introduced into superfluid helium by laser ablation of a metal targets immersed into liquid. Foils of chemically pure (99.99%) metals were used as targets. Indium–lead alloy has been prepared by heating the pieces of individual metals together in a sealed bulb to a temperature of 350 °C, well exceeded their melting points. After keeping at this temperature for 2 h the bulb was slowly cooled to 300 K. The quantized vortices nucleate at the focus of the pulsed laser beam as a result of the media heating, i.e., in the same place and at the same time as metal nanoparticles appeared. The product of the metal coalescence in the quantized vortex cores are the bundles of parallel-series of interconnected metallic nanowires attached to the tips of electrodes inserted into the condensation zone. By connecting the tips of neighboring electrodes the bundles closed the electrical circuit; distances between electrodes were 1.4 mm. Such a way the measurements of the electrical resistance were performed inside of cryostat. Part of the nanowires bundles were deposited on a carbon-coated copper grid placed on the bottom of the cell. After heating the cryostat up to 300 K, they were replaced to vacuum chamber of TEM electron microscope (JEOL JEM-2100) for studies of nanowire structure and morphology.

On heating, the relaxation of nanowire shape, which leads to their shortening, takes place at temperatures above 100 K [20]. This causes the breaks in the web formed by the nanowires, and, due to diminishing the percolation, the irreversible changes of the total resistance of the web of nanowires connecting the electrodes. For this reason we annealed the web by its heating up to a certain temperature Tan and subsequently cooled it down again. As a result the resistance value became reproducible for the temperatures below the annealing temperature, $T < T_{an}$. Typically, the temperature growth was achieved by a slow self-heating of whole cryostat. At low temperatures, reversible cycle "heating–cooling" was produced, under constant pumping of helium vapor, by a temporal irradiation of the sample immersed in liquid helium by thermal emission of incandescent lamps through the cryostat windows.

For normal metal nanowires as well as for superconducting nanowires, the diameter is one of the key factors defining the strength of all kinds of quantum size effects (QSE). But the origins of QSE for normal and superconducting metals are different: In normal metals the resistance is determined by the scattering of the conduction electrons on the surface of the wire or on the grain boundaries whereas in superconductors QSE may be due to a variety of reasons, including quantum phase slips and Coulomb suppression of superconductivity. Unfortunately, in experiment we cannot change the diameters of the nanowires because, for our nanofabrication method, the diameter is fixed for a given material. Instead we can produce nanowires out of different metals and alloys and compare them.

3. Results and discussions

We compare the temperature dependence of the resistance of the nanowire bundles made of three materials – niobium, platinum and indium–lead alloy. Fig. 1 shows the morphology and structure of the nanowires grown from niobium (a) and (b), indium–lead alloy (c) and (d) and platinum (e) and (f). All nanowires were of good quality, that is, without significant nodes and antinodes; their structure was close to being crystalline. It is seen from Fig. 1b, d, and f that the diameter of the nanowires for refractory niobium is about 4 nm, for the indium–lead alloy the diameter is about 8 nm, and the diameter of the platinum wire is 3 nm. The nanowire made of indium–lead alloy has no noticeable spatial inhomogeneity of the elemental composition, which is a consequence of the well-known fact that the alloy of these elements always forms a solid solution [21].