Superconducting transition of single-crystal tin microstructures

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Single-crystal superconducting microstructures have been fabricated. The resistances of tin whiskers were measured in a multiprobe configuration. Contacts were made of copper, gold, or niobium films using *e*-beam lithography followed by a lift-off process. Structures with normal metal probes showed unusual behavior: below the critical temperature of bulk tin, the resistance decreases in distinct steps and does not reach zero even when cooled down to 1 K. The origin of these phenomena is not clear but is likely related to a proximity effect.

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Metal microstructures and nanostructures are typically composed of thin films. This fabrication method results in samples being at the so-called ''dirty'' limit where the mean free path l is the smallest physical scale important for electron transport. At the dirty limit, the short mean free path leads to the renormalization of the superconductor coherence length $\xi(0)=0.85(\xi_0 l)^{1/2}$, which determines the dimensionality of the system. A long mean free path, resulting in a long coherence length, in single crystals expands the region close to the critical temperature T_c, over which the diverging coherence length $\xi(T)=\xi(0)(1-T/T_c)^{-1/2}$ governs the effective dimensionality of such a superconducting system. An additional advantage of pure single crystals is their superior homogeneity compared to structures at the dirty limit.

We have selected tin quasi-one-dimensional single crystals (whiskers) for fabrication of clean limit microstructures.[1] Typical cross sections and lengths of these objects are $\sim 1\ \mu\mathrm{m}^2$ and ~ 1 mm, respectively. Preliminary measurements showed a residual resistance ratio $(\mathrm{RRR})=R(300\,\mathrm{K})/R(4.2\,\mathrm{K})\sim 200$. Utilizing the value $\rho l=10^{-15}\,\Omega\,\mathrm{m}^2$, for the mean free path, one obtains $l \gtrsim 10\,\mu\mathrm{m}$ which is larger than the transverse dimension of the whisker. The mean free path in such an ultraclean limit, therefore, strongly depends on the surface properties.[2]

We used the whisker as the ''body'' of the microstructure and the probes were fabricated by conventional *e*-beam lithography (Fig. 1). Unfortunately, it appeared that a whisker freely lying on the Si substrate and being fixed only by evaporated thin film probes can not withstand the lift-off process. Crystals should be glued and the chip's surface should be sufficiently planar to allow lithography. AlliedSignal Inc. [3] spin-on-glass (SOG) was selected for planarization. Reactive ion plasma etching was used to remove the SOG from the top of the whisker. Afterwards, *e*-beam lithography was used followed by evaporation of microcontacts made of Cu, Au, or Nb. Typical widths of contacts were about 500 nm with thicknesses of about 200 nm. Spacing between voltage probes varied from 1.5–10.5 μm. Etching appeared to be the most crucial step, resulting in a low yield of ''measurable'' structures. From approximately 160 fabricated structures only 17 samples showed no obvious drawbacks on $R(T)$ dependencies and were selected for further measurements. These drawbacks were mainly large or infinite contact resistance between the electrodes and the whisker. We consider this effect is probably due to a combination of surface damage, while etching, and mechanical stress, while cooling, due to the mismatch of thermal expansion coefficients. The RRR of studied microstructures decreased from the initial value of about 200 down to 5–70.

A simple glass cryostat has been used for all experiments. The Earth's magnetic field was reduced by a μ-metal shield. Samples were immersed directly into the ^{4}He bath which was pumped down to 0.95 K with a diffusion booster. The majority of the measurements were made using a lock-in technique at a typical operating frequency of 19 Hz with Stanford Research System SR554, SR552, and SR560 front end preamplifiers, with noise levels of 110 pV/$\sqrt{\mathrm{Hz}}$, 2 nV/$\sqrt{\mathrm{Hz}}$, and 4 nV/$\sqrt{\mathrm{Hz}}$, respectively. The noise for our low ohmic samples is mainly determined by the total source impedance with a dominating contribution due to the whisker-probe contact resistance. However, independent of noise rejection capabilities of the devices, only samples with contact resistances of below a few tens of Ω at 4.2 K were subjected to further measurements. All experiments were done using a four-probe configuration.

Out of 17 ''successful'' microstructures, four were made using superconducting metal (niobium) for voltage and current probes. With these samples nothing unusual was observed. The superconducting transition was centered at the bulk critical temperature of tin, $T_c^{\mathrm{bulk}}=3.78\,\mathrm{K}$, and it's data plot had a rounded upper part which stretched to above 4.2 K. Probably, the shape of the superconducting transition can be attributed to the proximity effect due to a nearby superconductor with a higher critical temperature, which for our Nb films was about 7 K.

Half of the structures with normal metal probes (Cu or Au) showed a conventional superconducting transition at the critical temperature of bulk tin. However, the width of the transition was typically larger ($\sim$40 mK) than that based on prediction; that is, for the fluctuation governed form of the $R(T)$ dependence of a quasi-one-dimensional superconductor.[4] It is important to note, that such behavior was characteristic of samples which were measurable but had a higher contact resistance. Probably, the wide transition can be ac-

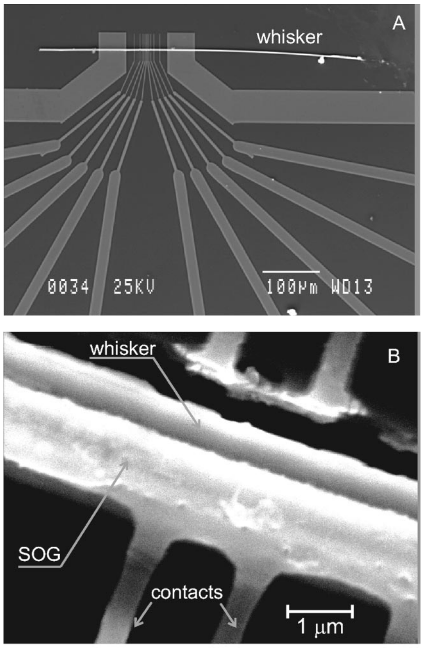

FIG. 1. (a) Scanning electron microscope image of one of the studied structures; (b) enlarged view of the contact region.

counted for by internal deformations due to stress while cooling since the crystal and the SOG have different thermal expansion coefficients.

The other half of the structures with normal metal probes had a rather low resistance: a few Ω between the corresponding voltage probes when measured using a two-probe method. We believe, that this low value stems from the quality of the lead's connection and it indicates that whisker-probe contact is Ohmic. For these samples, rather unexpected forms of resistance vs temperature $R(T)$ curves were observed (Fig. 2). The position of the top part of the ''superconducting transition'' corresponds to the critical temperature of bulk tin. The resistance does not drop to ''zero'' even when the temperature is lowered down to $T=1$ K. At the foot of the $R(T)$ dependence unusual steps develop. Within our experimental accuracy, these steps are reproducible even after several thermal cycling (Fig. 3). We believe, that the observed hysteresis of about 40 mK (Fig. 3) is not connected to any physical process in the measured samples. It originates from the space, of about 2 cm, between the sample and the thermometer, and from the temperature gradient within our long (110 cm) and narrow (30 mm) glass Dewar during cool-down and warm-up. Although measurement of each sample's $R(T)$ dependence takes more than a hour, still the temperature distribution inside the cryostat is not uniform.

A remarkable feature of this step structure is that the sample behaves as a ''multilevel system,'' at each plateau the signal is constant until the system ''switches'' to a new ''state'' (Fig. 3, inset). Between various samples, we could not find any universal dependence for the width (δT_{step}) and height (δV_{step}) of the steps. Nevertheless, for a given pair of voltage probes, at least, the heights of the steps look rather regular (Fig. 3). If to assume that each step corresponds to a superconducting transition of some part of a structure, than

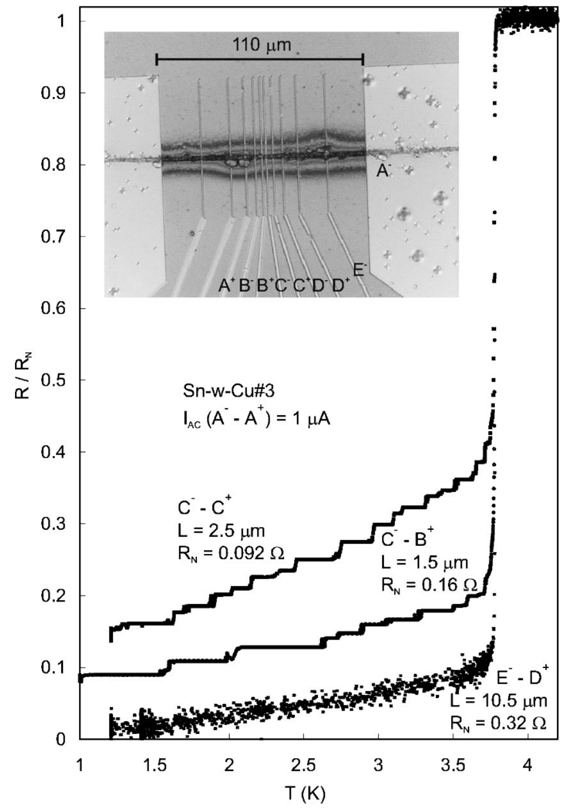

FIG. 2. Typical normalized $R(T)/R_N$ dependencies for various sections of the structure shown in the inset. Contact resistance for the probe pair E^{-}-D^{+} is larger than for other electrodes: the signal is noisy and the resistance drops to a lower value. Note that the normal state resistances R_N do not scale with the distance between the probes. Probably, this observation is related to the quality of the contact-whisker interface. The inset image shows the arrangement of the measurement probes.

one can estimate the average domain length which contributes to each step: $\delta L_{\mathrm{eff}}=20-900$ nm for various samples. It is most surprising that this effective length can be smaller than both the mean free path l and the coherence length $\xi(0)\approx 230$ nm. Probably, such a naive ''static'' calculation is not justified.

The application of a longitudinal magnetic field of a few mT causes only the top part of the $R(T)$ curve to shift (superconducting transition of bulk tin?) to a lower temperature but leaves the steps at the foot unchanged. The $R(T)$ dependencies were measured for a variety of ac currents, where the linearity of the $V(I)$ characteristics had been checked. In this limit, variation of the measuring current alters neither the shape of the step structure (Fig. 3, inset), nor the rest of the $R(T)$ dependency. Examples of the current-voltage characteristic $V(I)$ and its first derivative $dV/dI(I)$ are presented in Fig. 4. For all temperatures within the steplike resistive transition [Fig. 4(a), inset] the $V(I)$ dependencies are linear when currents are small ($I\leqslant 100\,\mu$A). At higher currents the current-voltage characteristics (Fig. 4) deviate from being linear, exhibiting broad transitions to states with higher dV/dI values but with smaller than normal state resistances R_N. The threshold current of the transition I_0 decreases with

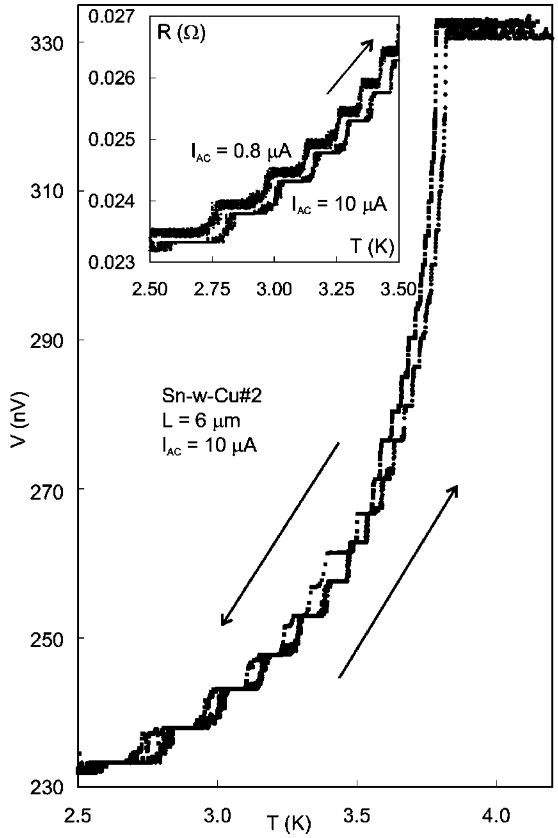

FIG. 3. An example of a pronounced steplike $V(T)$ dependence. Arrows indicate the direction of the temperature sweep. The small hysteresis is a consequence of the temperature measurement. For details see the text. The inset shows an enlarged view of several $R(T)$ steps from the low-temperature end of the same figure measured at two different currents. The curves are shifted slightly due to the zero offset drift of our preamplifier or lock-in amplifier.

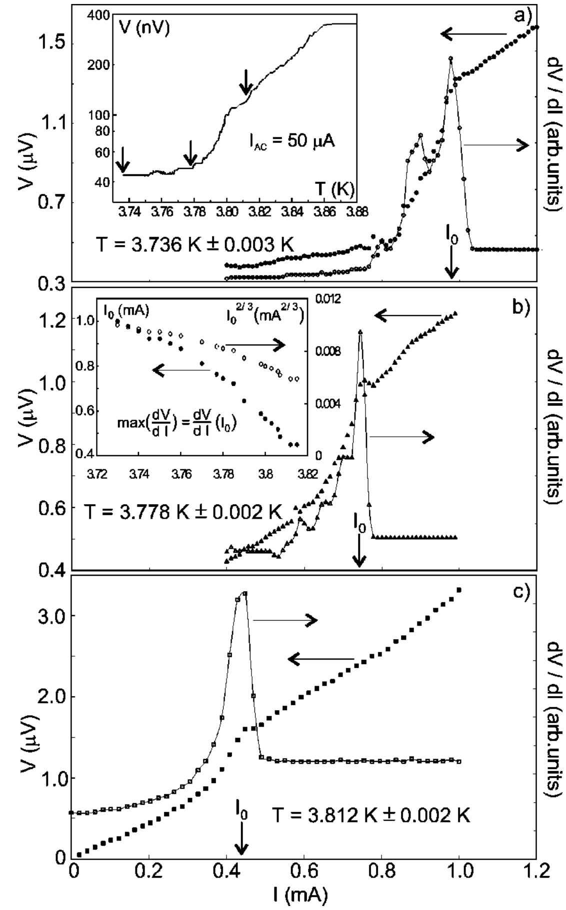

FIG. 4. Inset to (a): resistive transition of the structure with a distance of 8.8 μm between the probes and measured with a 50 μA ac bias current. (a)–(c): current-voltage characteristics $V(I)$ (solid symbols) and their first derivatives $dV/dI(I)$ (open symbols) taken at temperatures indicated by the arrows in the inset to (a). The dc current I was modulated with 10 μA ac component. The solid lines are guides for eyes. The $V(I)$ dependencies do not extrapolate exactly to the coordinate origin due to the drift of the zero offset of our dc multimeter. The temperature dependence of the threshold current I_0 is plotted in the inset to (b). For each temperature, the threshold current I_0 corresponds to the maximum of the derivative of the current-voltage characteristic.

temperature [Fig. 4(b), inset]. This tendency qualitatively resembles the well-known behavior of a critical current in a superconductor. However, the temperature dependence of the current I_0 does not follow exactly the "$\frac{3}{2}$ law" as expected for the critical current of a one-dimensional superconductor: $I_c^{1D} \propto T^{3/2}$ [Fig. 4(b), inset]. The origin of the nonlinearity of the current-voltage characteristics is not clear. Certainly, as the current-induced transition occurs from the state with finite resistance, the threshold current I_0 cannot be straightforwardly associated with superconducting critical current. We want to stress that all $R(T)$ steplike transitions [Figs. 2, 3, and 4(a), inset] were measured using ac currents with magnitudes much smaller than the threshold currents I_0. We believe that, in this linear limit, the process responsible for the steps on $R(T)$ dependencies is not related to the nonlinearity of the current-voltage characteristics, observed at much higher currents.

No correlation has been found between the distance of the voltage probes (sample resistance) and the parameters of the step structure (Fig. 2). However, the smaller the contact resistance, the more pronounced the step structure is. When exposed to the atmosphere at room temperature for a few days, the steps observed, on the same pair of contacts on the same structure, disappear: the transition becomes smooth and, within the measuring accuracy, a zero value of resistance is reached. Typically, after another few days at room temperature in normal atmosphere, the structures show very high contact resistance and cannot be measured further. We assume, that this degradation is due to aging of the whisker-probe contact. As a result of the plasma etching, the surface of the whisker is not smooth and the region under the probe contains small SOG islands [Fig. 1(b)]. Certainly, such a configuration favors continuous oxidation of the interface.

Similar steplike structures on $R(T)$ dependencies have been reported in early experiments with superconducting whiskers.[5,6] However, there are several principal differences in our observations. First, in those experiments, steps could be stimulated only by high dc current. At low currents, the transitions were rather smooth. In our case, steps were observed within a wide range of ac currents with much smaller current densities than reported in Refs. 5 and 6. Second, contacts to those samples were ''handmade'' by pressing the whisker into two superconducting islands of Wood's metal thus enabling two-probe measurements. In our case, steps were detected only in structures with normal metal electrodes. Note, that the steps on the I-V characteristics in these pioneering experiments[5] had a strong impact on the physics of nonequilibrium superconductivity; namely, current-induced activation of phase slippage. However, to our knowledge, the origin of the step structure on $R(T)$ dependencies has yet to be explained. Obviously, the orthodox model, dealing with fluctuations governed by thermal activation of phase slip centers,[4] is unable to explain the wide steplike $R(T)$ transitions in quasi-one-dimensional systems.

For the moment, we can only speculate on the origin of the steps in the $R(T)$ dependencies. We do not have an understanding of the accompanying effect: why the resistance does not drop to zero. In all samples displaying the step structure, within the accuracy of experiment, the $R=0$ point has not been reached down to $T=1$ K. Both phenomena have been observed in samples with the lowest contact resistance between the whisker and the normal metal probes. The effects disappear with the increase of the contact resistance. In the structures with superconducting electrodes, neither steps nor incomplete superconducting transition have been observed. One may suggest that the effects are related to the presence of a normal metal and its effect on the electronic properties of a superconductor in the clean limit.

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