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Controlled electromigration of thin Pb films started from the normal and from the superconducting state

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Abstract

We have investigated the electromigration of thin Pb films with a weak link, performing the main sample fabrication steps in ultra-high vacuum and avoiding organic chemicals. In the normal state, upon a current flow, joule heating leads to an increased temperature and electromigration proceeds due to thermal activation. Cyclic controlled electromigration at 300 K and at 77 K can be well-described by a model used previously where the feed resistance and the power dissipated in the contact remain constant during the thinning process with the new addition of a parallel resistance. This arises because the Pb film is not laterally confined. Controlled electromigration relies on current-heating a sample. This seems contradictory to superconductivity, because the superconducting current does not dissipate energy in the sample. Here we discuss how edge effects can be used to heat the sample first over the superconducting transition temperature and subsequently heating can be used to thin the sample. The superconducting transition of the thin film occurs at well-defined voltages, i.e. well-defined heating currents in the feeds. The same conductance steps during the superconducting transition are found in current–voltage measurements as well as during electromigration cycles. Due to a weak link in the thin film, the current density and the temperature distribution is not uniform in the sample, and therefore several conductance steps are found.

Keywords: electromigration, nanocontacts, superconducting transition, thin films, molecular electronics

1. Introduction

Controlled electromigration is a method to make nanometer-distance contacts that can be used to contact molecules [1–4]. The advantage of the method is that the contacts are planar, more stable compared to other methods and electrically driven. In principle the thinning method can be transferred to ultra-high vacuum conditions in order to avoid contaminations due to contact with air. If the method is combined with clean sample fabrication methods, where resists and other organic chemicals are avoided, one can be sure that only the molecule under study is contained in the sample. In addition, it is possible to fabricate many such contacts in parallel [5]. For molecular electronics a detailed knowledge not only of the molecule but also of the contacts is desirable. Such nanocontacts can be studied using the method of multiple Andreev reflections, a method for which superconducting contacts are needed [6]. Recently, electromigrated nanocontacts have been studied using this method and it was shown that the resistance state could be switched by electromigration between two values [7]. The method

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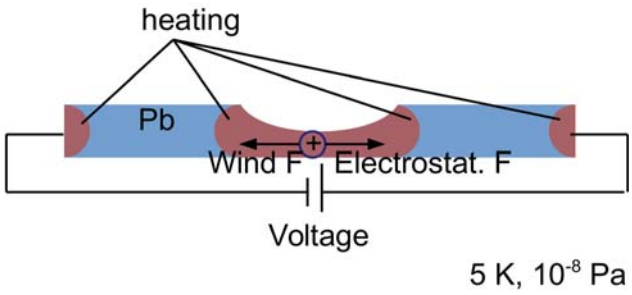

Figure 1. The superconducting transition is reached by heating from mechanical contact with the current-heated feeds. In the normal state, electromigration sets in at the thinned area where the contact is forming.

of multiple Andreev reflections together with calculations allowed to find information about the atomic configuration of the different resistance states. Therefore it is desirable to study nanocontacts of superconducting metals.

Here, we study electromigration thinning using a cyclic process started at temperatures above and below the superconducting transition temperature of Pb thin films. We demonstrate that the main fabrication steps can be performed without the use of organic chemicals and in ultra-high vacuum. For controlled electromigration joule heating is required that is suppressed in the superconducting state. Above the transition temperature electromigration thinning can be well-described using a model where the feed resistance and the power dissipated in the forming nanocontact is constant. We add a parallel resistance to the model to account for the relatively wide Pb film. Below the superconducting transition temperature of Pb we study the interplay of superconductivity and electromigration. If we employ the same cyclic process as at higher temperatures, we first observe heating above the superconducting transition temperature resulting from joule heating the feeds, see figure 1. Only after reaching the normal state the Pb film is thinned due to controlled electromigration. The method allows to study how the sample reaches the superconducting transition. The current density varies for different parts of the sample due to varying film thicknesses and thus several superconducting jumps are reproducibly observed.

2. Experimental methods

For the electromigration process, we first prepared Pb thin films samples by shadow evaporation. In this way, we avoid contamination of the sample by organic chemicals.

For sample preparation first the substrate, naturally oxidized Si, was mounted on a sample holder base plate and a mask was fixed in front of the substrate. For some samples in addition a wire was mounted in front of the mask to create a thinner area of the sample i.e. a weak link as a starting point for electromigration. Then the substrate was introduced into the ultra-high vacuum chamber and cleaned by heating using a constant current through a heater located at the base of the sample holder. Temperatures above 100 °C were reached. After that the substrate was cooled by liquid nitrogen and Pb was deposited on it through the mask using an electron beam evaporator. The film thickness was determined using the ionic current of the evaporated Pb measured during evaporation. The ionic current had been calibrated using a quartz crystal microbalance. Subsequently the sample was removed from the vacuum chamber, contacted with graphite glue or silver paste and 100 μm-thick Cu-wire, and re-introduced into our physical properties measurement system (PPMS, Quantum Design, California, USA) or re-introduced into the same ultra-high vacuum chamber for electromigration. Since Pb oxidizes in air, we minimized the time necessary for contacting. After re-introduction to the ultra-high vacuum chamber the sample was cleaned again by a similar heating procedure reaching temperatures above 100 °C. Several hours of heating were necessary until the base pressure was reached again due to outgassing of the glue used for contacting.

3. Results and discussion

Prior to electromigration, the resistance of a 60 nm-thick Pb film was measured in the PPMS in a four-terminal configuration as a function of temperature and magnetic field, see the supporting information (available at stacks.iop.org/JPhysD/50/185301/mmedia). We found a residual resistance ratio (RRR), $R(T = 300\,\mathrm{K})/R(T = 8\,\mathrm{K})$ of 20 in reasonable agreement with the value of 50 obtained for a 90 nm-thick Pb film on glass [8] or the value of 14 obtained for a 200 nm-thick Pb film also on glass [9]. The transition width is on the order of 0.03 K. This shows that our Pb films are of a good structural order. The superconducting phase transition in zero field occurred at $T_c = 7.2$ K. Bulk Pb is a type-I-superconductor with $T_c = 7.2$ K. In the thickness range above 10 nm this value remains unchanged. Below 250 nm, i.e. in the thickness range used here, the thin film geometry supports flux vortices in the thin film for geometrical reasons, i.e. Pb becomes a type II superconductor. By investigating the linear dependence of the upper critical magnetic field H_{c2} on temperature, we find the electronic mean free path $\ell = 15$ nm and confirm that the assumption of the dirty limit applies, see the supporting information. In addition, we calculate the product of resistivity and mean free path $\rho_0 \cdot \ell$ and obtain 1.9 fΩm^2, compared to the value found in literature of 1.5 fΩm^2, see [8]. We believe that the difference is due to oxidation since the oxidation leads to a reduced conductive film thickness and thus to an artificially large value of the resistivity. This effect should be stronger for our samples compared to the reference [8] since our film thickness was much smaller.

When a current runs through a metal, the metallic ions are subject to electromigration forces, i.e. the Coulomb force and the collective force generated by the flow of electrons and their collisions with the ions, the wind force. In this way, the thermally activated diffusion of the ions obtains a preferential direction. This process depends critically not only on the current density, but also on temperature, because diffusion is strongly temperature-dependent. The process of electromigration is self-accelerating: if the metallic contact is thinned, the resistance and thus the local temperature increases, leading

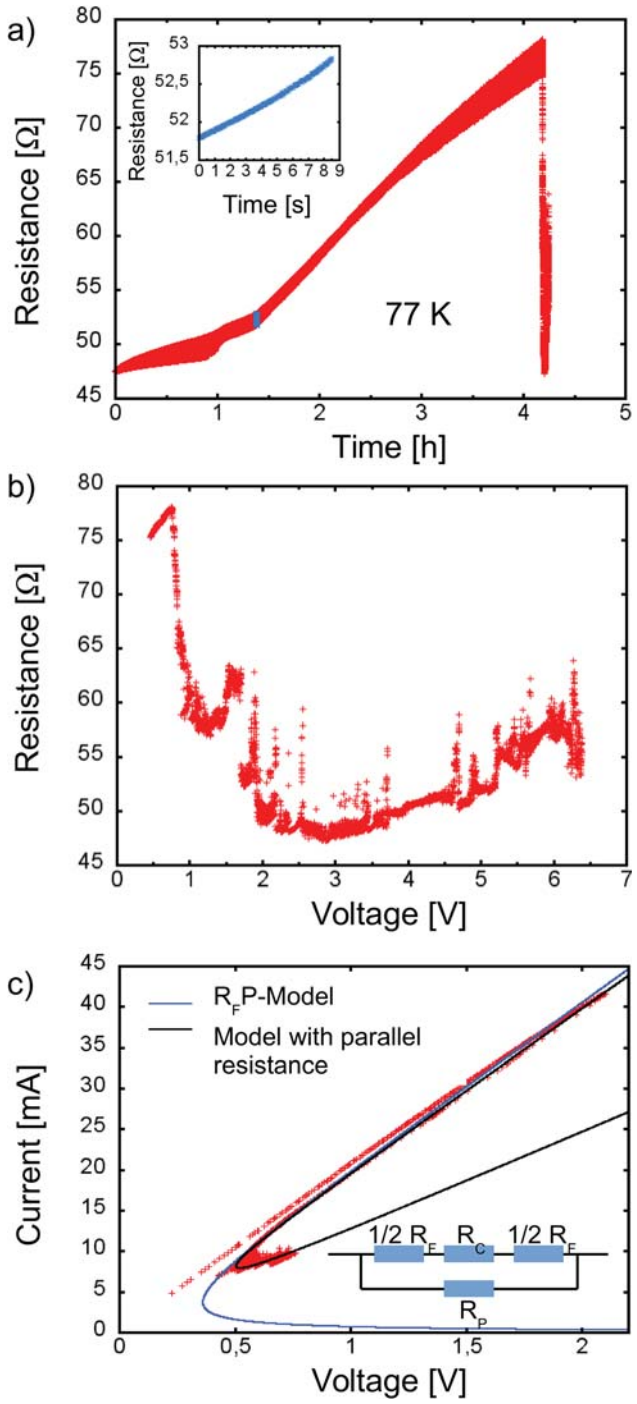

Figure 2. (a) Total dataset of the resistance acquired during cyclic controlled electromigration of a Pb thin film at 77 K. The resistance increases in general before the sample is destroyed after a sudden decrease of the resistance. The data of one single electromigration cycle has been marked in blue. Inset: the data of the single electromigration cycle marked in blue is enlarged. (b) Irregular resistance trace of the sample prior to destruction. (c) If the data is plotted in the $I(V)$ diagram, a RFP model with additional parallel resistance can be used to describe the data as described in the text. Blue curve: RFP model without parallel resistance ($R_F = 49\ \Omega$ and $P^* = 0.67$ mW). Black curve: RFP model with parallel resistance ($R_F = 50\ \Omega$, $P^* = 0.5$ mW and $R_P = 32\ \Omega$).

to enhanced electromigration. This process can lead to overheating and destruction of the contact. In order to control electromigration [1–3], we have defined a computer-controlled process in which we increase the voltage in two terminal configuration, until a pre-chosen value of the resistance is reached both due to heating and electromigration. Then we automatically reduce the voltage and start a new cycle where the pre-chosen resistance value is automatically increased compared to the previous cycle. The process is stopped when a pre-chosen final resistance value is reached. With a similar process a continuous and permanent resistance increase is obtained from one cycle to another in Au [4] and Cu [10].

When we subject a 140 nm-thick Pb film to controlled electromigration at 77 K, we obtain a continuous and permanent increase of the resistance from cycle to cycle for 4 h (figure 2(a)). The increase of the resistance within one cycle is shown in blue color and in the inset. Then suddenly there is a resistance drop followed by an irregular behavior of the resistance as a function of voltage (figure 2(b)). This could be caused by melting. As a result, the pre-chosen resistance value is not reached and the voltage is automatically increased up to 6.3 V as compared to a maximal voltage of 0.7 V in the previous cycle. At 6.3 V the sample was destroyed.

For Au and Cu the end points of the cycles have often been described successfully by a model where the feed resistance R_F and the dissipated power P^* in the contact are assumed to remain constant [3]. In this model the feed resistance includes most of the thin film wire and the terminal resistance. Since the contact resistance $R_c = P^*/I^2$ is a function of current, the voltage of the end points becomes hyperbolic

$$U = R_F I + P^*/I. \tag{1}$$

Sometimes, this process is disturbed because the feed resistance suddenly increases and the dissipated power decreases [11]. This can be attributed to contact formation and further confinement of the heated area. In this case, the data can be described by using several hyperbolas. However, for the data of the Pb film (figure 2(c)), even several hyperbolas do not suitably describe the data in particular for the low current and high voltage regime. To describe the data better, we introduce a parallel resistance R_P (see the inset of figure 2(c) for a sketch of the model), and obtain

$$U = \frac{R_P}{2}\left[\left(2\frac{R_F}{R_P} + 1\right)\cdot I \pm \sqrt{I^2 - 4\frac{P^*}{R_P}}\right]. \tag{2}$$

Using $R_F = 50\ \Omega$, $P^* = 0.5$ mW and $R_P = 32\ \Omega$ gives a suitable description of the data. The values are reasonable compared with similar data obtained previously from Au and Cu samples [4, 10]. The additional parallel resistance is thought to occur in the Pb layer. Previous studies of the structure of electromigrated nanocontacts show that controlled electromigration leads to slit formation with parts of the slit only nanometers apart [11]. It has been shown that heating leads to a resistance drop that was attributed to thermal expansion of the metal and partial closure of the slit [12]. We assume that during slit formation, part of the thin film is still electrically connected while another part is already interrupted by the slit. The parallel resistance represents the part of the Pb film that is still electrically connected.

This behavior is similar to what we have observed for electromigration of a 125 nm-thick Pb film at room temperature.

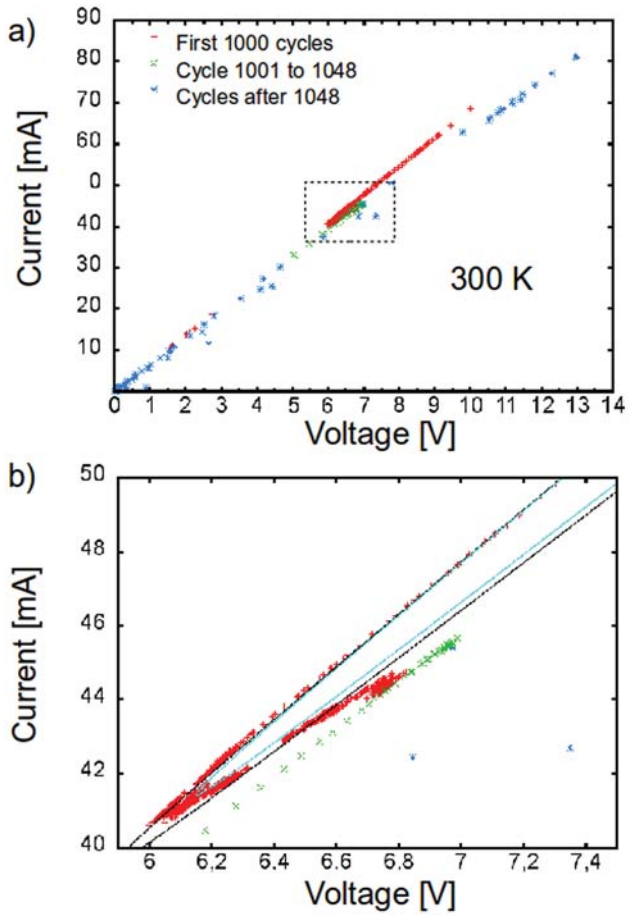

Figure 3. (a) $I(V)$ diagram of the data acquired during electromigration of a Pb thin film at room temperature. Most of the data is measured between 6 and 7 V, therefore this area is enlarged. In (b) RFP models with parallel resistance are shown in black ($R_F = 145\ \Omega$, $P^* = 3$ mW and $R_P = 7.6\ \Omega$, fit using Levenberg–Marquardt algorithm) and blue lines ($R_F = 145\ \Omega$, $P^* = 3$ mW and $R_P = 7.0\ \Omega$, fit by eye).

After 20 h of electromigration the cycles become irregular and result in a sudden jump of the resistance to a value around 1 kΩ. The irregular behavior could be caused by melting. For such an irregular behavior, it is likely that the protocol leads to destruction of the sample. Also for this sample we show the data in the $I(V)$ diagram (figure 3(a)) and observe that 900 of 1000 data points occur in the window between 6 and 7 V. Enlarging this part of the diagram (figure 3(b)) shows that the cycle end points are well-described by the model with parallel resistance introduced above with $R_F = 145\ \Omega$, $P^* = 3$ mW and R_P values between 7.0 and 7.6 Ω.

We then attempted to study controlled electromigration starting the process at sample temperatures below the superconducting transition of Pb. The full data set for a 100 nm-thick sample is shown in figure 4(a)). Here, the sample is heated through the feeds. The feed resistance (8 Ω) can be read from figure 4(c)) when the sample is in the superconducting state, i.e. from the resistance value of the gray curve at 0.2 V. A marked jump is observed in the data of figure 4(a)) at around 0.5 V. A closer view (gray curve in figure 4(c)) shows that in the area between 0.25 and 0.55 V, several resistance jumps are observed. These are reproducible about 50 times. Due to their reproducibility the jumps cannot be explained by electromigration and are identified as superconducting jumps. Their presence and reproducibility shows that in this part of the process, the sample is unaltered by electromigration. Starting from cycle number 182, the full superconducting jump is observed whereas in the cycles before, only partial switching is observed. In the normal state, i.e. for the data above 0.7 V in figure 4(a)), electromigration proceeds but cannot be described by the model discussed above in a simple manner, since the last cycle, the blue curve in figure 4(a), reaches larger voltage and current values compared to intermediate cycles.

After 1000 cycles a current–voltage measurement is performed (figure 4(b)). At the start of the measurement, the sample is still cooling down as the voltage is lowered from values above 2 V to values below 0.3 V. As a result of cooling down, the sample switches from the normal state to the superconducting state as can be seen from the jump to higher current values at $V = 0{,}3$ V in the red curve. The current–voltage characteristics shows that the main superconducting transition occurs at voltages of 0.5 V. At this voltage, the heating power from the feeds is sufficiently large to reach the superconducting transition temperature. In figure 4(c) we compare these values with the ones obtained during electromigration cycles and find that the superconducting transition is always found at similar voltages. Some hysteresis occurs because the precise voltage at which the transition occurs depends on the history of the sample and of its temperature determined by the heating history. Also we observe a smaller jump in the same voltage range in the electromigration cycles as well as in the $I(V)$ characteristics. We believe that this jump is related to the geometry of the sample with a thinner area comprised between two weak links as shown in figure 5(a)).

To create a weak link in the center of the thin film, on some samples we used a Cu wire of a width of 50 μm mounted on the mask. Figure 5(b)) shows a typical sample obtained using this method. Areas where the Pb is thin appear green in the optical image. It becomes clear that in the center of the position of the wire, there is full shadow, therefore the sample shown in figure 5(b)) had no electrical contact. To establish electrical contact, on other samples we used two evaporation steps. Here, figure 5(a)) we discuss the sample preparation used for the sample generating the data shown in figure 4. First, only 20 nm were evaporated, then the sample was moved to another position in the vacuum chamber and later 77 nm of Pb were evaporated. The position of the mask had slightly changed between the two evaporation procedures. Two weak links were formed as visible in figure 5(a) taken after electromigration. One of the weak links had 20 nm film thickness and the other 77 nm film thickness. Since the green area on the right hand side is enlarged compared to the projection of the wire onto the sample plane, we believe that this area was affected by electromigration. The small area between the two weak links, where the half-shadows of the two evaporation processes overlapped, could be responsible for the smaller one of the two superconducting jumps in figure 4(c).

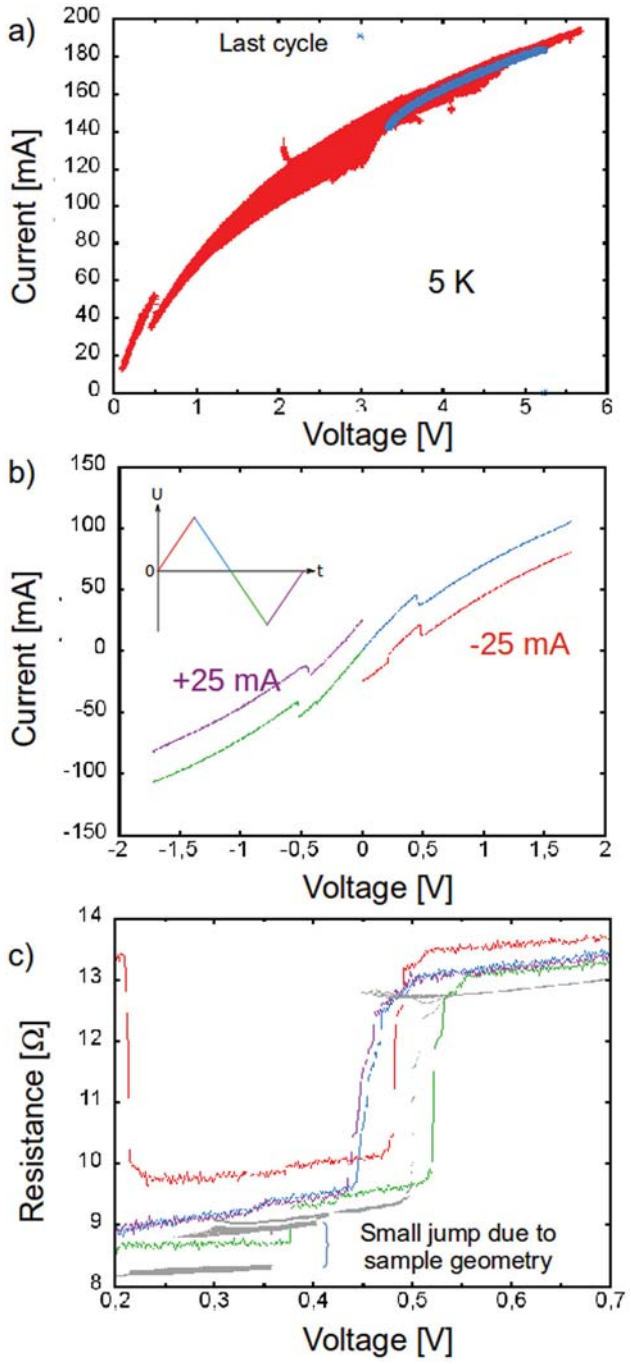

Figure 4. (a) Total data points measured during electromigration of a Pb thin film with weak links at 5 K. Below 0.5 V several superconducting jumps are observed. After 1000 cycles, a current–voltage characteristic is measured and leads to a small peak due to cooling at 2 V. The data from this current–voltage measurement is shown in (b): the red curve follows immediately to an electromigration cycle with increasing voltage. Since the electromigration cycle leads to heating of the Pb thin film, the sample cools down during the start of the red curve. This is visible through the superconducting jump at approximately 0.2 V. Then follows the blue curve with decreasing voltages. After that increasing negative voltages are applied (green curve). For the purple curve, the negative voltages are decreased in amplitude. In (c) the resistance data calculated from the current–voltage characteristics (red, blue, green, purple as in part (b), for the green and the purple curve the voltage scale is multiplied by −1) are compared to the superconducting transitions observed during the electromigration process (gray). The small jump is believed to result from a small area of the sample where the film thickness was thinner as explained in the text.

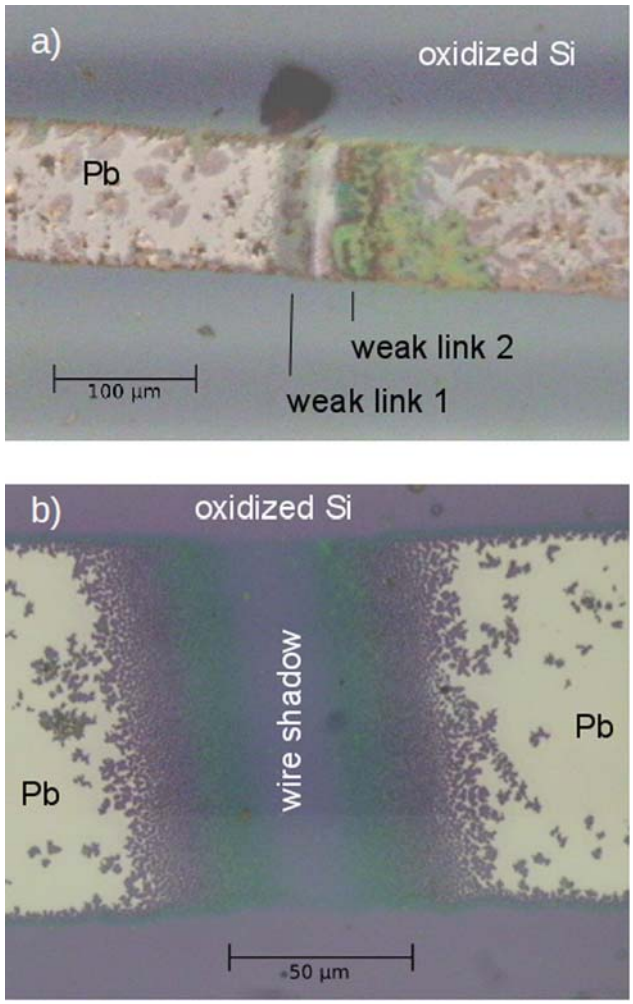

Figure 5. (a) Sample investigated in figure 4 at 5 K after electromigration. The sample shows two weak links because a wire was placed in front of the evaporation mask, Pb was deposited, then the sample was moved, then another layer of Pb was deposited. (b) A test sample with only one evaporation step for comparison.

The area near the two weak links has a locally increased resistance in the normal state compared with the other parts of the thin film due to its reduced film thickness. Due to this reduced film thickness, the local current density is increased in this area. We estimate the critical current of the thinned area to be 1.1 A and the critical current for the other parts of the thin film to 5.6 A (see supporting information for details). This implies that the superconducting transition is reached by heating rather than by reaching the critical current since we observe the transition at currents of 43 mA and 53 mA. This is related to the sample geometry. However, the calculated values for the critical current are the ones valid for $T \ll T_c$, whereas here the region close to T_c is relevant where the critical current approaches zero. In this region, there is a small temperature regime where the critical current of the thinner area is indeed smaller than the measured current, causing the jump of smaller height in figure 4(c). This could be viewed as an effective reduction of the critical temperature in the part of the film with reduced film thickness. For only one weak link we expect the position in voltage and the relative height of the two superconducting jumps to differ from what is observed here.

To test this hypothesis we measure the height of the smaller superconducting jump with respect to the larger one and obtain a relation of 1:6. If we estimate the resistance of the thinner sample area with respect to the other parts of the film, we obtain a relation of 1:15. The total resistance of the thinner sample area is smaller due to its much smaller length. Hence it causes the smaller superconducting resistance jump. Even taking imprecisions in the measurements into account, the comparison between the two ratios implies that this cannot fully explain the small jump. Possibly, the smaller superconducting area could be affected by oxidation in a stronger way than the other parts of the sample as would be the case for an oxide layer of uniform thickness.

We compare our results to the ones shown previously in literature: Luo *et al* [13] have investigated Pb contacts made by electron beam lithography with a width of 500 nm and a bow-tie structure with 100 nm width at its smallest width. They performed electromigration using a single voltage ramp at 4.2 K. The formation of a nanocontact is identified in scanning electron microscopy images taken before and after electromigration. This shows that the gap obtained is larger than the one usually obtained for controlled electromigration, where often the gap cannot be observed in scanning electron microscopy due to its small size and lack of resolution [4]. However, the definition of a weak link by confining the thin film in the lateral direction (i.e. using a smaller width) is advantageous compared to the method of reducing the thickness used here because this could suppress parallel conduction through the Pb. Luo *et al* observed conductance jumps on the order of the conductance quantum and the excess current below the superconducting gap due to Andreev reflection.

In [7] cyclic reproducible resistance jumps have been observed while the sample remained in the superconducting state. This is in contrast to our observations where we attribute reproducible jumps due to the superconducting transition. However, there are several differences between the experiment described in [7] and the one described here: in [7] the current direction is periodically reversed, reversing the electromigration direction and the movement of atomic positions while here we keep the same current direction. In addition in [7] the contact is much smaller compared to the one desribed here and comprises only a few atoms. If the jumps observed here were due to atomic rearrangements, we would expect even more jumps at higher voltages, while we observe less jumps at higher voltages.

The results are in line with our own observations on Au thin films [14]: we have investigated the local structure of wide Au thin film samples subjected to a similar electromigration process compared to the one used here. The formation of a slit across the thin film in the direction perpendicular to the current flow and extensions in the direction of the current flow have been observed. The conclusion was drawn that several hot spots are formed at the locations of the extensions, hundreds of nanometers away from the slit. If a similar process occurred in Pb thin films, local heating at the extensions could cause melting of the Pb film on a scale large enough to affect the slit and close it by molten material. For macroscopic Cu wires we have found previously that oxidation during controlled electromigration in air enhances the stability of the wires during the electromigration process while it was difficult to thin down the same wires to atomic dimensions in vacuum [10] in line with the results shown here.

4. Conclusion

We have investigated electromigration in Pb thin films by employing a cyclic controlled electromigration process. Samples are fabricated avoiding the use of organic chemicals, and performing the most important fabrication steps in ultra-high vacuum. For temperatures of 77 K and 300 K, i.e. for temperatures above the superconducting transition temperature of Pb, we observe a standard electromigration process known from other materials with the new addition of a parallel resistance. This resistance occurs because we do not laterally confine the Pb thin film. When we subject the Pb film to the same cyclic process at temperatures below the superconducting transition of Pb (at 5 K), we observe for low voltages applied during the first cycles that electromigration remains inactive because no energy is dissipated in the superconducting part of the sample. The voltage is gradually increased by the controlled electromigration procedure. At larger voltages joule heating in the feeds leads to heating of the superconducting film to the transition temperature. The voltage for which the transition temperature is reached is well-defined and corresponds to a certain heating current and a certain temperature reached in the Pb film due to heating from the feeds. This voltage differs for different parts of the sample due to varying effective superconducting transition temperatures caused by varying film thicknesses. We study the superconducting transition during the cyclic process as well as during current–voltage measurements and reproducibly obtain the same transition states.

Acknowledgments

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