Anomalous magnetoresistance oscillations and enhanced superconductivity in single-crystal Pb nanobelts

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Using atomically flat single-crystal Pb ultrathin films grown on Si(111) substrate by molecular beam epitaxy, we fabricated Pb nanobelts of 28 atomic monolayers thick, 285 nm wide, and 10 μm long by focus ion beam technique. The transport property is measured and the results are compared to the Pb films. We find that below the superconducting transition temperature T_C, the resistance of the nanobelts exhibits clear and reproducible oscillations as a function of perpendicular magnetic field, which are not observed in the Pb films. In addition, T_C enhancement was also observed in the nanobelts. The results suggest that the single-crystal Pb nanobelts prepared on Si semiconductor substrate offer a great opportunity for integrating superconducting circuits in a single Si chip. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2945280]

Superconductivity in quasi-one-dimensional (quasi-1D) nanowires is one of the most intriguing problems in condensed-matter physics.[1–6] Many interesting physical phenomena, such as thermally activated phase slip,[7–9] quantum phase slip,[10,11] superconductor-insulator transition,[12] and proximity effect,[13,14] were observed in quasi-1D systems as lately developed experimental techniques have enabled the fabrication of superconducting wires with a diameter reaching to certain degree of the 1D limit.[15,16] Magnetic field lifts the time reversal symmetry of the spin and orbital states of paired electrons and suppresses the superconducting transition;[17] many interesting results such as magnetoresistance oscillations were also observed under a magnetic field.[7,18–20] However, the nanowire samples used in these studies were either amorphous or granular, or their sizes are ill defined. To understand these interesting behaviors, single-crystal superconducting systems with controlled dimensionality are highly desirable.

Here, we report on a method to fabricate single-crystal Pb nanobelts with nanometer scale control of their sizes by using combined molecular beam epitaxy (MBE) thin film growth and focus ion beam etching techniques. These Pb nanobelts exhibit enhanced T_C and reproducible resistance oscillations as a function of magnetic field perpendicular to the plane of the sample below T_C, which were not found in the Pb films of the same thickness from which the nanobelts were made, and are also different from previous reports.[18,20]

The experiments were performed in an ultrahigh vacuum low-temperature scanning tunneling microscopy (STM) system with a MBE chamber for *in situ* preparation of thin films (Unisoku USM1300). The Pb thin films used for fabrication of nanobelts were prepared on high resistivity Si(111) substrates, which are insulating at low temperature.[21] The details were reported elsewhere.[22–27] Figure 1(a) shows a typical STM image of the atomically smooth Pb films with a thickness of 28 atomic monolayers (MLs). To fabricate nanobelts, the film was deposited with 4 ML Au as a protection cover[22] and was transferred out of the UHV growth chamber. The

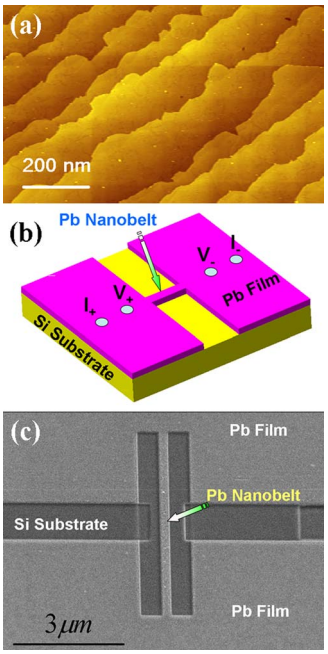

FIG. 1. (Color online) (a) A scanning tunneling microscope image of the 28 ML atomically smooth Pb thin film on the Si(111)7×7 substrate. The image was recorded at 80 K with a constant current of 100 pA. (b) The schematic graph for the transport measurement across the Pb nanobelt. The four contact electrodes are made of indium. (c) A scanning electron micrograph (SEM) of the Pb nanobelt made of the Pb film shown in (a). The Pb nanobelt is 28 ML thick, 285 nm wide and 10 μm long. The dark region on the two sides of the Pb nanobelt is exposed Si surface, which isolates two blocks of the Pb film. The dimension in the length direction of the nanobelt is compressed since the SEM picture was taken at the 52° tilted sample stage in a FIB system.

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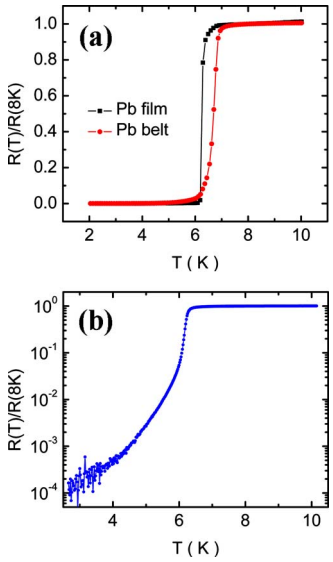

FIG. 2. (Color online) (a) Resistance as a function of temperature measured from the Pb film in Fig. 1(a) and the Pb nanobelt in Fig. 1(c), respectively. The vertical scale is normalized to the resistance at $T=8$ K. The transition temperature T_C of the Pb nanobelt is obvious higher than the T_C of the Pb film. (b) Logarithm resistance vs temperature of a 300 nm wide and 23 ML thick Pb nanobelt. Below T_C, the resistance tail is obvious.

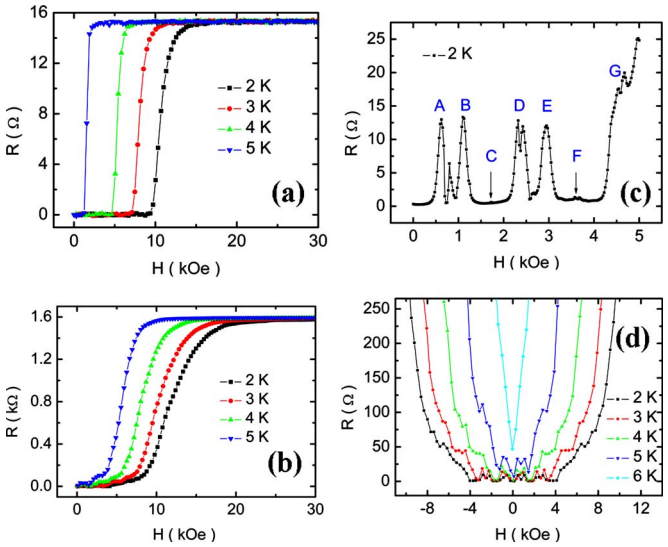

FIG. 3. (Color online) (a) Magnetoresistance of the Pb film in Fig. 1(a) with a magnetic field applied perpendicular to the film at different temperatures. (b) Magnetoresistance of the Pb nanobelt in Fig. 1(c) with a magnetic field applied perpendicular to the film at different temperatures. (c) Close view of (b) in low magnetic field regime at 2 K for clarity. (d) Magnetoresistance oscillations of the Pb nanobelt in Fig. 1(c) at 2, 3, 4, 5 and 6 K, respectively, with both positive and negative perpendicular fields.

nanobelts were then made by a commercial focused ion (Ga) beam etching system (FEI-DB235), as schematically shown in the Fig. 1(b). The etching current was set less than 10 pA to minimize contamination and structure damage by the Ga ions. Four indium electrodes with Au wires of 25 μm in diameter were made and connected to the surface of the two parts of the film [see Fig. 1(b)] for transport measurements. Since the thickness of the Pb films can be precisely controlled, the nanobelt shown in Fig. 1(c) exhibits well-defined dimensionality and is of single crystal except for trivial damage of its long edge sides by the ions, as compared to the amorphous and granular samples previously studied.[15,18,20]

The Pb nanobelt shown in Fig. 1(c) is 28 ML (~8 nm) thick, 285 nm wide, and 10 μm long, and the equivalent cross section is about 48×48 nm^2, which is comparable to the BCS coherent length of bulk Pb at 0 K, $\xi_0^{\text{bulk}}=90.5$ nm (Ref. 28) and the coherence length of ~27 nm in the Pb film [estimated by $\xi\approx(\xi_0 l)^{1/2}$ at the dirty limit,[25] where l is the mean free path for the film]. Please note that the Pb nanobelt is 285 nm wide, while the size of the vortex core (which is the same as the coherence length) in the Pb film is only 27 nm. Thus vortices can enter into the belt and the Pb nanobelt is not 1D or quasi-1D superconductor.[29] However, with a very small thickness-to-width ratio (<0.03) the single-crystal Pb nanobelt shows different behavior with the Pb film and displays some characteristics of 1D superconductor, as discussed below. 1D behavior in quasi two-dimensional samples was reported in disordered superconducting NbN nanowires and wide superconducting strips with a hole in the middle.[30,31]

Figure 2(a) shows R-T curves of the 28 ML Pb film [see Fig. 1(a)] and the Pb nanobelt [see Fig. 1(c) for comparison]. Both the film and nanobelt show nearly ideal superconducting transition and almost zero residual resistance, indicating high quality of the samples. We also note that the onset T_C (6.9 K) of the Pb nanobelt is obviously higher than that (6.3 K) of the Pb film. The enhancement of T_C was verified in another three samples. We know that the T_C of the ultrathin single-crystal Pb film is lower than the T_C of the bulk Pb.[22–27,32] However, from Pb film to Pb nanobelt, the T_C is increased.

One may think that the enhancement of T_C is related to the enhanced surface electron-phonon scattering.[33,34] However, the surface area in the nanobelt is increased only by 0.2% compared to the thin film case, which is not enough to explain the increase of 9%. Currently, we have no satisfied explanation of this observation.

In order to study the R-T property of the Pb nanobelt at low temperature, the logarithm of the resistance versus temperature for a 300 nm wide and 23 ML thick Pb nanobelt is plotted in Fig. 2(b). We can see clear nonlinear resistance tail below T_C. The curve is smooth and shows a positive curvature, as one expect from the models with quantum phase slips in superconducting nanowires.[10] The behavior was confirmed in other samples.

Figure 3(a) shows the resistance of the 28 ML film as a function of the magnetic field (H) applied perpendicularly to the film at different temperatures (below T_C). The film shows a sharp superconducting transition and almost zero residual resistance. We can see that below T_C the upper critical field $H_{C\perp}$ of the superconducting Pb film gradually decreases with increasing temperature. Figure 3(b) shows the corresponding R-H curves of the Pb nanobelt. The critical field $H_{C\perp}$ of the superconducting nanobelt gradually decreases with temperature; however, the superconducting transition region becomes wider. It is noted that the normal state resistance of the Pb film is ~15 Ω and the Pb nanobelt is ~1.6 kΩ in the normal state. The Pb film is about 2 mm wide and the distance between the two voltage electrodes is about 0.7 mm. According to the film resistance (15 Ω), we can derive that the resistance of the nanobelt should be about 1.5 kΩ, which is close to our measurement result (1.6 kΩ). Furthermore, below the $H_{C\perp}$ of the Pb nanobelt, the magnetoresistance exhibits some oscillations in superconducting state.

To further understand the magnetoresistance behavior of the superconducting Pb nanobelt, we show the *R-H* curve of the Pb nanobelt in low magnetic field regime at 2 K (below T_C) in Fig. 3(c). An oscillation in magnetic resistance is evident. At zero field, the resistance is close to zero (less than 1 Ω). With increasing field, the resistance remains small and reaches peak "A" at 0.6 kOe at which another resistance of 13 Ω appears. With further increasing field, the resistance oscillates. From the "A" to "F," we can see an oscillating period of 0.6 kOe although the "C" and F are suppressed and the periodicity is not well defined. In addition, from the "D," "F," and "G" double peaks are present. Due to the transition at $H_{C\perp}$, the "G" deviates the oscillation period. Above 5 kOe, the resistance increases continually and the oscillations disappear gradually. Figure 3(d) shows magnetoresistance oscillations at various temperatures below T_C in both positive and negative perpendicular fields. This figure gives evidence that the oscillation in the Pb nanobelt is reproducible (not just some sort of noise) and might be considered quasiperiodic. The big oscillation amplitude of more than 10 Ω can be exploited to fabricate nanoscale SQUID-like[18] devices and on-off functional and logical nanodevices.

The observed magnetoresistance oscillation is not understood well currently. It seems that there are mesoscopic superconducting ringlike structures in the Pb nanobelt, which might induce the periodic magnetoresistance oscillation in superconducting regime, such as Little–Parks-like[35] oscillations. We note that although the oscillation is reproducible for different samples, the period of oscillation is not well defined. A possible explanation is that some normal regions might occur in the nanobelt or in the films connected to the nanobelt in the presence of a magnetic field. These regions can block the current and cause a random resistance fluctuation, as reported previously by Prozorov *et al.*[36] In this case, the magnetoresistance oscillations cannot be periodic.

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