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An unusual magnetoresistance effect in the heterojunction structure of an ultrathin single-crystal Pb film on silicon substrate

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Abstract

Superconductor films on semiconductor substrates have drawn much attention recently since the derived superconductor-based electronics have been shown to be promising for future data processing and storage technologies. By growing atomically uniform single-crystal epitaxial Pb films of several nanometers thick on Si wafers to form a sharp superconductor–semiconductor heterojunction, we have obtained an unusual magnetoresistance effect when the Pb film is superconducting. In addition to the large fundamental interest in this effect, the simple structure, and compatibility and scalability with current Si-based semiconductor technology offer a great opportunity for integrating superconducting circuits and detectors in a single chip.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The use of dissipationless superconducting components will produce denser and faster chips since the resistance of the interconnecting metal circuits is a major source of heat generation and charging time [1]. Motivated by rapid progress in superconducting electronics [2–5], such as logic circuits, sensitive detectors and nonvolatile memories, superconductor–semiconductor hybrid structures have become an attractive field in recent years [6–10]. A 'holy grail' would be an integration of the superconducting films with Si-based microelectronic technology.

Here, we report our experimental observation of an unusual magnetoresistance phenomenon in simple Pb–Si superconductor–semiconductor heterojunctions. By growing ultrathin single-crystal Pb films (<10 nm in thickness) on Si(111) substrates, a giant negative magnetoresistance effect was observed when the Pb thin films become superconducting at low temperature.

2. Experimental procedure

Our Pb thin films were prepared on heavily doped n^+ Si(111) substrates by the standard molecular beam epitaxy (MBE) technique [11]. During growth, the Si substrates were cooled down to 95 K by liquid nitrogen (LN_2) to achieve atomically smooth single-crystal Pb thin films, as reported elsewhere [12–18]. Figure 1(a) shows a typical scanning tunneling microscopy (STM) topographic image of the Pb thin film with a thickness of 26 atomic monolayers (ML), from which the atomically smooth nature of the film is immediately evident. Before the samples were taken out from the ultra-high vacuum growth chamber for transport property measurement, 4 ML Au was deposited on the film to protect it from contamination and surface oxidation in ambient conditions [12, 16]. The transport measurements were carried out by using the standard four-electrodes method in a physical property measurement system (Quantum Design Model 6000).

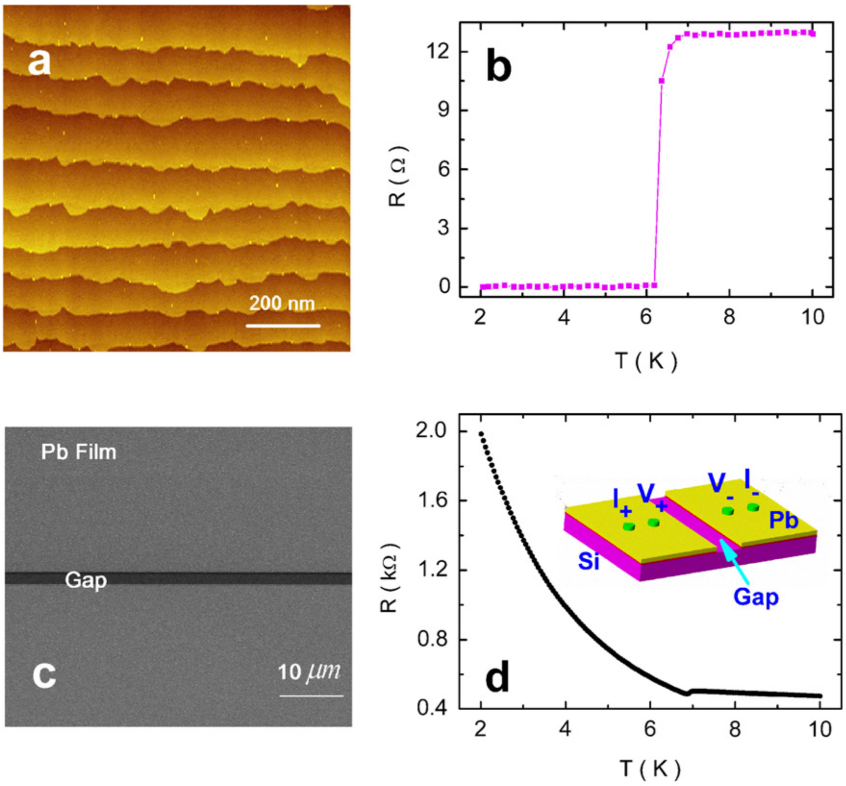

Figure 1. (a) A scanning tunneling microscope image of the 26 ML atomically flat Pb thin film. (b) R versus T curve measured from the Pb film shown in (a), showing a superconductivity transition at a temperature of 6.4 K. (c) A scanning electron micrograph of the Pb film after a 2 μm wide gap (the dark region) was fabricated. (d) R versus T obtained from the Pb–Si–Pb double-junction structure. The inset is the schematic graph for the transport measurement across the Pb/Si(111) heterojunctions.

As shown in figure 1(b), the film exhibits a superconducting transition at 6.4 K (T_C), and no residual resistance was found.

To measure the transport properties through the Pb–Si heterojunctions, the film was cut into two parts with a 2 μm wide gap made by a focused ion beam (focused ion beam etching & depositing system, FEI-DB235) (see figure 1(c)). The etching current with Ga ions was less than 10 pA, and when the gap was not long enough to separate the two parts of the Pb film, the sample showed almost the same transport property shown in figure 1(b). Therefore, contamination and damage of the structure by the Ga ions could be mostly avoided. Four indium electrodes with Au wires of 25 μm in diameter were made and connected to the two parts of the film and the measurement geometry is schematically shown in the inset of figure 1(d). Because the resistances of both the doped Si substrate and Pb film are very small (the resistance of the n^+ Si wafer used is below 0.1 Ω even at 2.5 K), the measurement mainly reflects the transport property of the two Pb–Si heterojunctions. Figure 1(d) shows the resistance–temperature (R–T) curve of this double-junction structure. Below 7.0 K the resistance drops slightly at first. Then, with further decrease in temperature, the resistance increases rapidly.

3. Results and discussion

Figures 2(a) shows the measured resistance (R) as a function of the magnetic field (H) applied perpendicularly to the Pb film at different temperatures. It is clear that the resistance decreases rapidly (at an average rate of $\sim$0.42 Ω Oe^{-1} for $T = 2.5$ K) with increasing magnetic field, eventually reaching a plateau at a critical field H_C. The curve is symmetric under both positive and negative fields, forming a resistance peak around zero field. It is also clear that both the maximal resistance and H_C increase with decreasing temperature. Figure 2(b) is a close-up view of figure 2(a) near zero magnetic field for clarity. The vertical scale is normalized to the resistance at zero magnetic field. At $T = 5.5$ K, the resistance decreases by a factor of 1.3 when H is increased from 0 to 0.9 kOe. Remarkably, that factor increases to 3.1 with a field change of 2.6 kOe at $T = 2.5$ K, in comparison with a factor of about 2 for the traditional giant magnetoresistance (GMR) effect in the Fe/Cr system under a field of 20 kOe at 4.2 K [19]. Besides the large peak, the resistance also exhibits a weak minimum at a magnetic field just below H_C. The resistance minima at the positive and negative fields are approaching a zero field and become more pronounced with increasing temperature (below 7 K). The phenomenon was verified on several samples. We found almost the same result when we used a diamond cutter to separate the Pb film. Therefore, the Ga ion etching process in the experiment did not affect the observed behavior.

For comparison, the R–H scan of the 26 ML Pb film is shown in figure 2(c). From this figure one can clearly see that, at the same temperature, the upper critical field H_{C2} of the Pb film is much larger than the H_C of the Pb–Si junctions (see figures 2(a) and (b)). For example, at 2.5 K, the H_{C2} of

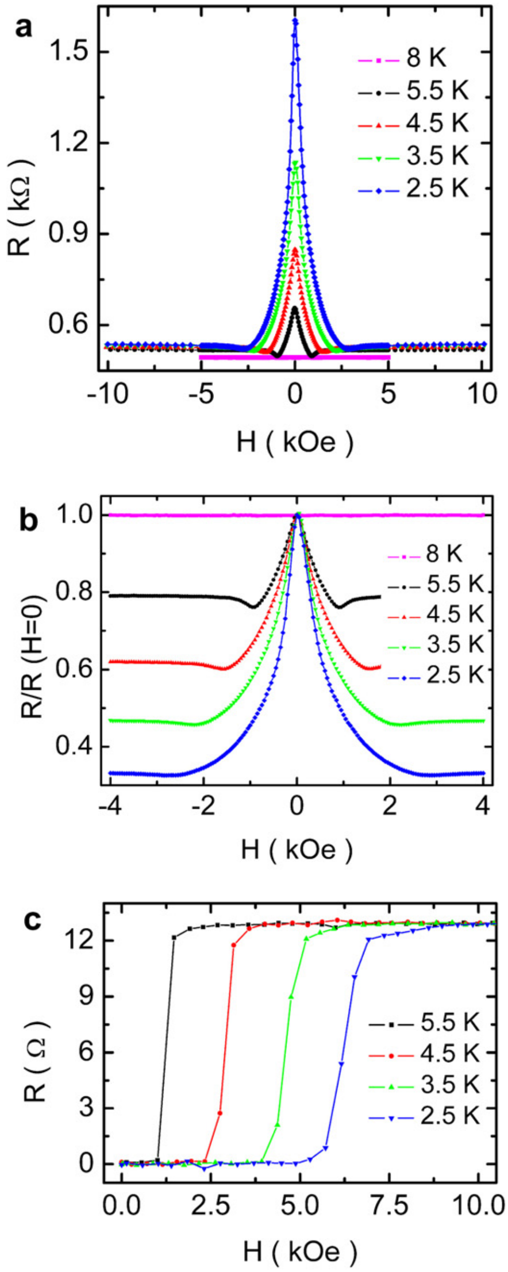

Figure 2. (a) Magnetoresistance of the heterojunctions with a magnetic field perpendicular to the film at different temperatures. (b) Close-up view of (a) near zero magnetic field for clarity. The vertical scale is normalized to the resistance at zero magnetic field. Note that there is no GMR effect when the film is in the normal state (pink line). (c) R versus H curves of the 26 ML Pb film with a magnetic field perpendicular to the film at indicated temperatures.

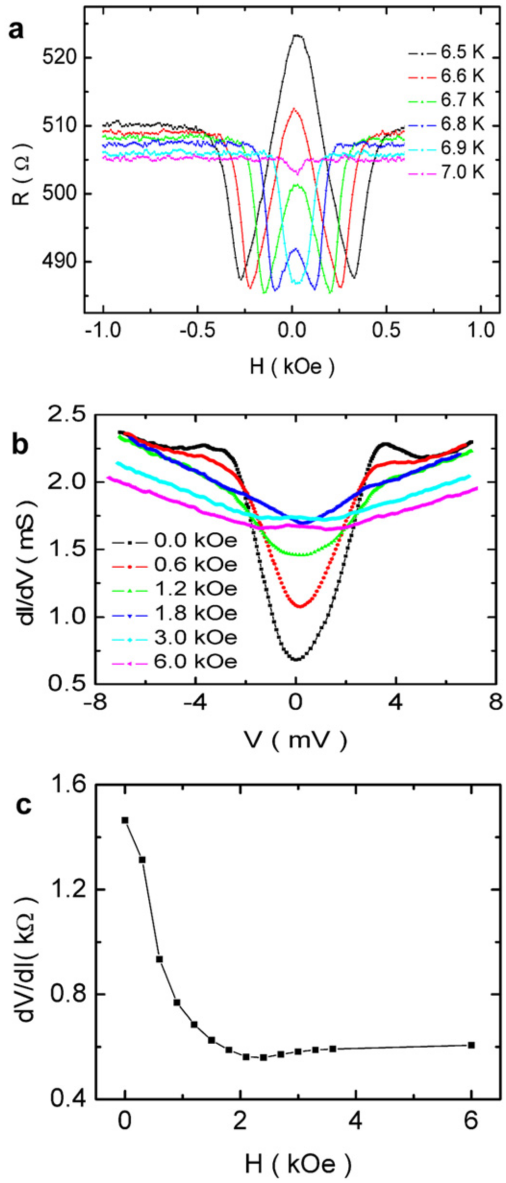

Figure 3. (a) R–H curves of the heterojunctions with a magnetic field perpendicular to the film at 6.5, 6.6, 6.7, 6.8, 6.9 and 7.0 K, respectively. (b) Differential conductance dI/dV versus voltage V curves of the heterojunctions at indicated magnetic fields at 2.5 K. (c) dV/dI versus H curve of the heterojunctions at 2.5 K when the applied voltage is zero. The data are from dI/dV–V curves.

the Pb film is 7 kOe, while the corresponding H_C of the Pb–Si junctions is only 2.8 kOe. It is surprising that between 7 and 2.8 kOe, although the Pb film is still superconducting, the negative magnetoresistance effect no longer exists.

Figure 3(a) shows more details of the R–H curves of the Pb–Si junctions. Sharp valley-like resistance minima are found from 6.5 to 6.8 K. With increasing temperature, the large resistance peak at zero field gradually fades away and the valley-like resistance minima are approaching the zero field. Above 6.7 K the resistance peak becomes lower than the plateau in high field, and at 6.9 K the peak basically disappears and the resistance minima at positive and negative fields are merged together around zero field. The valley-like resistance minimum almost disappears at 7.0 K and the R–H curve becomes a straight line above 7.0 K. The data clearly reveal that the resistance of the Pb–Si junctions is very sensitive to the temperature when the temperature is below 7.0 K. A very small change (0.1 K) in temperature could induce more than a 10 Ω change in resistance. Such sensitivity makes the Pb–Si junction highly promising for mass production of new cryogenic temperature detectors on Si chips [4, 20]. Furthermore, the behavior that the resistance increases quickly after dropping and finally becomes constant with an increasing field may find application in future functional devices. Please notice that the R–H curves at 6.5 and 6.7 K were obtained by scanning the field from -1 to 1 kOe while the data at 6.6 K were obtained with a scanning field from 1 to -1 kOe. We can see that the positions of the resistance peaks at 6.5 and 6.7 K are the same. But, compared to the peak at 6.6 K there is a small shift of 20 Oe. We attribute the peak shift to the trapped vortices in the ultrathin Pb films. Due to the pinning of some vortices, the magnetoresistance curves show small hysteresis with different field scan directions.

In order to further understand the effect of the superconducting Pb film on the observed magnetoresistance effect, differential conductance experiments were carried out with the same sample in one week. Figure 3(b) shows the $\mathrm{d}I/\mathrm{d}V$–V curves measured under various magnetic fields at 2.5 K. At zero field, the differential conductance forms a large valley around zero voltage, which gradually disappears as the magnetic field is raised. The shape of the curve is reminiscent of the tunneling result for a superconductor–insulator–normal metal structure [21]. However, our transport result is from a Pb–Si–Pb structure, which is not an ideal tunneling system for the measurement of the superconducting gap in a superconductor. In figure 3(c), we plot the differential resistance versus the magnetic field at zero voltage, which is obtained from the data of $\mathrm{d}I/\mathrm{d}V$–V in figure 3(b). It is clear that the differential resistance behavior is almost the same with the direct R–H measurement (figures 2(a) and (b)), which further confirms our negative magnetoresistance phenomenon.

Recently, negative magnetoresistance in disordered thin films and wires has also been observed [22–25]. However, the enhanced negative magnetoresistance behavior found in heterojunctions of a single-crystal ultrathin superconductor film and semiconductor substrate has never been reported before. Since the shape of the $\mathrm{d}I/\mathrm{d}V$–V curve at zero field looks like the tunneling spectra of the ultrathin Pb films fitted with BCS-like DOS [21], one possible source of the unusual magnetoresistance behavior is the electronic tunneling in a superconductor–normal metal (S–N) junction [26], namely, the tunneling between the superconducting Pb film and the n^+ Si substrate through the Schottky barrier [27] at the epitaxial Pb/Si(111) interface. With an increasing magnetic field, the electron density for tunneling increases. Thus the S–N-like tunneling is enhanced with increasing field. Accordingly, the resistance of the junction decreases with increasing field. Nevertheless, this simple picture does not explain the finding that the H_C of the structure is much less than the upper critical field H_{C2} of the Pb film. It is also difficult to understand the fact that the resistance exhibits a weak minimum at H just below H_C.

Another possible qualitative explanation is from the BTK model [28] and Andreev reflection [29]. We know that there is a tunneling barrier between the Pb film and the Si substrate due to formation of the Schottky barrier at the interface. Because the resistance of the sample is not too large (above 2 K, the resistance is below 2 kΩ, see figure 1(d)), we believe that the strength of the barrier is intermediate (between zero barrier and a strong tunnel barrier). Blonder *et al* (BTK) [28] introduced a δ-function potential barrier of strength Z at the interface to study the electric tunneling. If the strength Z is zero, there is no barrier at the interface between the normal and the superconducting metals. The electrical current transfer process is a novel reflection process described by Andreev [29]. This situation applies to a normal metal–superconductor junction. If the Z is very large (for example, larger than 10), there is a classic high barrier tunneling junction and electron tunneling dominates the electron transport. For our sample, Z is not zero, but is not large either. According to the BTK model, for this situation, the probability of Andreev reflection is increased when the electron energy is changing from 0 to $\Delta(T)$ (2Δ is the superconducting gap according to BCS theory [30]). Then it decreases rapidly with a further increase in electron energy. Since the effective superconducting gap decays with increase of an applied magnetic field, the electron energy in our measurement becomes close to Δ with increasing field. Accordingly, the probability of Andreev reflection increases and the resistance decreases. When the electron energy equals Δ, the probability of Andreev reflection reaches a maximum value: correspondingly, the resistance reaches a minimum in the R–H curves. However, the BTK model and Andreev reflection cannot explain the fact that we got almost the same magnetoresistance behavior by using 50 and 500 nA currents for the measurement, since at zero field the energy of the electron for the 500 nA measurement current is 0.8 meV at 2.5 K, corresponding to 0.08 meV for the 50 nA current. It is also not easy to understand the H_C of the structure is much less than the upper critical field H_{C2} of the Pb film.

4. Conclusion

We do not as yet have a satisfactory model to explain the unusual magnetoresistance effect found in the Pb–Si structure. The Pb–Si interface formed by epitaxy and the formation of quantum well states, which greatly modulates the electronic structure near the Fermi energy [12–14, 16, 31] in the present Pb film, may also play important roles. We expect that our work will stimulate further theoretical and experimental studies.

The unusual magnetoresistance effect in superconductor–semiconductor heterojunctions may be utilized in developing a magnetic field controlled 'on–off' device or a high-sensitivity field sensor. Because it is from the electron transport across the Pb/Si(111) interface, fabrication of any devices based on the effect could be scaled up for mass manufacture using the well-established microelectronics technology. This effect may also be utilized, or need to be avoided, in future hybrid circuits of the emerging superconducting electronics [32].

Acknowledgments

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