## Negative magnetoresistance in fractal Pb thin films on Si(111)

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Using a low temperature method, the authors have prepared atomically flat Pb ultrathin films on Si(111)-7×7 surface. Room temperature annealing of the films results in a percolation morphology with fractal vacancy islands where the Si substrate is exposed. The fractal film with a nominal thickness of 23 ML exhibits enhanced onset superconducting transition temperature of 7.0 K and negative magnetoresistance with wide magnetoresistance terrace under perpendicular magnetic field when the film is in superconducting state. They attribute the phenomena to the coexistence of two superconducting phases in this fractal film. © 2007 American Institute of Physics. [DOI: 10.1063/1.2712511]

One of the most interesting problems in condensedmatter physics is superconductivity in reduced dimensionalities. As an ideal testing ground of low dimensional superconductivity, two-dimensional (2D) superconducting films have attracted much attention recently.<sup>1-6</sup> Indeed, in amorphous or granular 2D superconductor films (including onedimensional case), negative magnetoresistance and other anomalous behaviors have been found.<sup>5,7–9</sup> The negative magnetoresistance effect appears to be a signature of quantum phase transition or interplay of disorder. From the point view of technological applications, the films<sup>10–12</sup> and related nanodevices<sup>13–17</sup> have been predominately prepared or fabricated on semiconductor substrates. Along this line, Pb on Si(111) probably represents the most interesting system because of the strong quantum confinement effects that lead to various intriguing phenomena such as "magic film thickness," "preferential island heights," and "oscillating super-conductivity transition temperatures."<sup>18–21</sup>

In this letter, we report on transport property of Pb thin films with a unique fractal-like morphology grown on Si(111)-7×7 surface. Under the perpendicular magnetic field, the films exhibit distinctive negative magnetoresistance effect with wide magnetoresistance terrace. Structure analysis by scanning tunneling microscopy and electron microscopy reveals that the observed effect originates from the coexistence of two superconducting phases in the system.

Our Pb thin films were prepared on Si(111)-7×7 surfaces by standard molecular beam epitaxy technique.<sup>22</sup> To achieve atomically flat single crystal Pb thin films over a macroscopic area, the Si substrates were cooled down to ~95 K by liquid nitrogen (LN<sub>2</sub>) during growth, as reported elsewhere.<sup>19,20</sup> Figure 1(a) shows a typical scanning tunneling microscopy topographic image of the Pb thin film of a thickness of 23 ML; the atomically smooth nature of the film is immediately evident. After depositing 4 ML Au on the film as a protecting cover,<sup>19</sup> the sample was transferred out of the UHV chamber for transport property measurement by the standard four electrodes method on a physical property measurement system (Quantum Design, model 6000). Four indium electrodes with Au wires of 25  $\mu$ m in diameter were made and connected to the surface of the film. As shown in Fig. 1(b), the sample exhibits a sharp superconducting transition at 6.1 K ( $T_c$ ), and no residual resistance was found.

When the film stays at room temperature in atmosphere for 48 h, the morphology of flat film becomes fractal-like, as shown by the scanning electron microscopy image in Fig. 1(c). Figure 1(d) shows the resistance-temperature (R-T)curve of the sample. The normal state resistance is much



FIG. 1. (Color online) (a) Scanning tunneling microscope image (500  $\times$  500 nm<sup>2</sup>) of the 23 ML atomically flat Pb thin film. (b) *R* vs *T* curve measured from the Pb film in (a), showing a sharp superconductivity transition at 6.1 K. (c) Scanning electron micrograph of the fractal-like Pb film. The dark regions are the Si substrate and the gray and white ones are the Pb film. (d) *R* vs *T* curve of the fractal-like Pb film shown in (c).

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FIG. 2. Magnetoresistance of (a) the fractal-like Pb film and (b) the single crystal Pb film with a magnetic field applied perpendicular to the film, measured at the temperatures of 2 and 3 K, respectively.

FIG. 3. (Color online) (a) Magnetoresistance of the fractal-like Pb film at different temperatures. (b) Close view of (a) near zero magnetic field for clarity.

larger than that of the continuous flat film. With decreasing temperature, the resistance increases and reaches a maximum at 7.0 K. Below 7.0 K, the resistance drops rapidly, reaches a minimum at 5.4 K, and then increases exponentially. We notice that the onset temperature  $T_C$  (7.0 K) in Fig. 1(d) is higher than the  $T_C$  (6.1 K) of the Pb film in Fig. 1(b).

Figure 2(a) shows the resistance (*R*) measured at a temperature of 2 K as a function of the magnetic field (*H*) applied perpendicularly to the film. It is clear that when the magnetic field is smaller than 2.5 kOe, the sample shows an obvious negative magnetoresistance effect. 2.5 kOe is a characteristic critical field  $H_1$ , above which the resistance increases rapidly with field. It reaches another quick increase regime at another critical field  $H_2$  (16.9 kOe) after a wide resistance terrace (~11.9 kOe) and eventually saturates at 21.5 kOe. For comparison, *R*-*H* scan of the flat film [Fig. 1(a)] measured at 3 K is shown in Fig. 2(b), which indicates that there is only one perpendicular critical field  $H_C$  and no residual resistance is found below  $H_C$ .

The *R*-*H* curves of the fractal film at different temperatures are shown in Fig. 3. The negative magnetoresistance effect becomes weaker at higher temperature. At the same time, the characteristic field  $H_2$  drops rapidly and eventually merges with  $H_1$  at 5 K. Above 7.0 K, the superconductivity vanishes.

As mentioned before, negative magnetoresistances in disordered thin films and wires have been reported by several groups.<sup>5,7–9</sup> For quench-condensed ultrathin amorphous Bi films, a parallel-field negative magnetoresistance was observed in the immediate vicinity of the thickness-tuned superconductor-insulator transition,<sup>7</sup> which was attributed to be a signature of quantum fluctuations of the order parameter associated with quantum critical point. In the case of granular Pb thin films, the large negative magnetoresistance was Downloaded 31 Aug 2007 to 166.111.26.232. Redistribution subject to the s

believed to be dominated by electron tunneling between the superconducting grains.<sup>5</sup> A common feature in these studies is that the negative magnetoresistance effect always appears with superconductor-insulator transition. In the present case, the samples are of single crystal in nature despite the fractal morphology and are still conducting (with a resistance  $<500 \Omega$ ) when the negative magnetoresistance takes place. Besides, the two critical fields observed have not been reported before either. Superfluid density fluctuations could lead to negative magnetoresistance in amorphous wires when the temperature is close to the superconductor-insulator transition.<sup>9</sup> Some magnetoresistance terraces were also observed. However, the model cannot be applied here since the negative magnetoresistance and wide resistance terraces occur at the temperatures lower than the  $T_C$ .

The anomalous magnetoresistance effect observed here can be understood qualitatively as follows. Owing to the fractal-like structure of the Pb film, two kinds of superconducting phases, related to the percolation Pb structures and the flat 2D Pb islands [Fig. 1(c)] should contribute to the total resistance observed. In the low temperature regime, electron tunneling dominates the transport property of the percolation structures. When a magnetic field is applied, the superconducting gap is suppressed. Reduction of the gap will lead to a significant decrease of the resistance, which we believe is the origin of the negative magnetoresistance. From Fig. 1(c), we can see that the percolation structures (white) are thicker than the flat 2D island parts (gray). Hence, a higher critical field  $(H_2)$  is expected for the flat parts while a lower one  $(H_1)$  for the percolation structures. Their difference defines the width of the magnetoresistance terrace which can be controlled by the morphology. So far we do not have a definite picture for the enhancement of the transition temperature  $T_{C}$ . A very speculative explanation is that it is related to the enhanced surface electron-phonon scattering, since the surface area is significantly increased when the film becomes fractal.<sup>23,24</sup>

In summary, negative magnetoresistance has been observed in fractal-like Pb single-crystalline films. The effect originates most likely from the interconnecting wires when they are at superconducting state. The two characteristic critical fields can be attributed to the coexistence of the two kinds of superconducting phases in the system: the percolation structures with lower  $H_c$  and the 2D islands with higher  $H_c$ . The effect shows both fundamental interest and possible application in fabrication of hybrid devices based on the traditional microelectronics and the emerging superconducting quantum electronics.<sup>25,26</sup>

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<sup>1</sup>John M. Blatt and Colin J. Thompson, Phys. Rev. Lett. **10**, 332 (1963).

- <sup>2</sup>B. G. Orr, H. M. Jaeger, and A. M. Goldman, Phys. Rev. B **32**, 7586 (1985).
- <sup>3</sup>Wenhao Wu and P. W. Adams, Phys. Rev. B 50, 13065 (1994).
- <sup>4</sup>A. Frydman, O. Naaman, and R. C. Dynes, Phys. Rev. B **66**, 052509 (2002).
- <sup>5</sup>R. P. Barber, Jr., Shih-Ying Hsu, J. M. Valles, Jr., R. C. Dynes, and R. E. Glover, III, Phys. Rev. B **73**, 134516 (2006).
- <sup>6</sup>Kevin A. Parendo, K. H. Sarwa, B. Tan, and A. M. Goldman, Phys. Rev. B **73**, 174527 (2006).

- <sup>7</sup>Kevin A. Parendo, L. M. Hernandez, A. Bhattacharya, and A. M. Goldman, Phys. Rev. B **70**, 212510 (2004).
- <sup>8</sup>N. Markovic, A. M. Mack, G. Martinez-Arizala, C. Christiansen, and A. M. Goldman, Phys. Rev. Lett. **81**, 701 (1998).
- <sup>9</sup>P. Xiong, A. V. Herzog, and R. C. Dynes, Phys. Rev. Lett. **78**, 927 (1997).
- <sup>10</sup>J. Eroms, D. Weiss, J. De Boeck, G. Borghs, and U. Zulicke, Phys. Rev. Lett. **95**, 107001 (2005).
- <sup>11</sup>C. Castellana, F. Giazotto, M. Governale, F. Taddei, and F. Beltram, Appl. Phys. Lett. 88, 052502 (2006).
- <sup>12</sup>F. Giazotto, P. Pingue, and F. Beltram, Mod. Phys. Lett. B 17, 955 (2003).
- <sup>13</sup>Y. Makhlin, G. Schon, and A. Shnirman, Nature (London) **431**, 138 (2004).
- <sup>14</sup>J. L. O'Brien, G. J. Pryde, A. G. White, T. C. Ralph, and D. Branning, Nature (London) **426**, 264 (2003).
- <sup>15</sup>P. K. Day, H. G. LeDuc, B. A. Mazin, A. Vayonakis, and J. Zmuidzinas, Nature (London) **425**, 817 (2003).
- <sup>16</sup>G. Wendin and V. S. Shumeiko, Science **292**, 231 (2001).
- <sup>17</sup>R. Held, J. Xu, A. Schmehl, C. W. Schneider, and J. Mannhart, e-print cond-mat/0506647 (2005).
- <sup>18</sup>Tai-Chang Chiang, Science **306**, 1900 (2004).
- <sup>19</sup>Y. Guo, Y. F. Zhang, X. Y. Bao, T. Z. Han, Z. Tang, L. X. Zhang, W. G. Zhu, E. G. Wang, Q. Niu, Z. Q. Qiu, J. F. Jia, Z. X. Zhao, and Q. K. Xue, Science **306**, 1915 (2004).
- <sup>20</sup>Y. F. Zhang, J. F. Jia, T. Z. Han, Z. Tang, Q. T. Shen, Y. Guo, Z. Q. Qiu, and Q. K. Xue, Phys. Rev. Lett. **95**, 096802 (2005).
- <sup>21</sup>Mustafa M. Ozer, James R. Thompson, and Hanno H. Weitering, Nat. Phys. **2**, 173 (2006).
- <sup>22</sup>J. L. Li, J. F. Jia, X. J. Liang, X. Liu, J. Z. Wang, Q. K. Xue, Z. Q. Li, J. S. Tse, Z. Y. Zhang, and S. B. Zhang, Phys. Rev. Lett. **88**, 066101 (2002).
- <sup>23</sup>Mingliang Tian, Jinguo Wang, James S. Kurtz, Ying Liu, and M. H. W. Chan, Phys. Rev. B 71, 104521 (2005).
- <sup>24</sup>D. G. Naugle, J. W. Baker, and R. E. Allen, Phys. Rev. B 7, 3028 (1973).
- <sup>25</sup>A. J. Leggett, Science **296**, 861 (2002).
- <sup>26</sup>Aharon Kapitulnik and Guy Deutscher, Phys. Rev. Lett. **49**, 1444 (1982).