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Modifying Quantum Well States of Pb Thin Films via Interface Engineering *

FU Ying-Shuang(付英双)[1,2], JI Shuai-Hua(季帅华)[1,2], ZHANG Tong(张童)[1,2], CHEN Xi(陈曦)[2], JIA Jin-Feng(贾金锋)[2], XUE Qi-Kun(薛其坤)[2], MA Xu-Cun(马旭村)[1]**

[1]*Institute of Physics, Chinese Academy of Sciences, Beijing 100190*
[2]*Department of Physics, Tsinghua University, Beijing 100084*

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We demonstrate the importance of interface modification on improving electron confinement by preparing Pb quantum islands on Si(111) substrates with two different surface reconstructions, i.e., Si(111)-7 × 7 and Si(111)-Root3×Root3-Pb (hereafter, 7 × 7 and R3). Characterization with scanning tunneling microscopy/spectroscopy shows that growing Pb films directly on a 7 × 7 surface will generate many interface defects, which makes the lifetime of quantum well states (QWSs) strongly dependent on surface locations. On the other hand, QWSs in Pb films on an R3 surface are well defined with small variations in linewidth on different surface locations and are much sharper than those on the 7 × 7 surface. We show that the enhancement in quantum confinement is primarily due to the reduced electron-defect scattering at the interface.

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Confinement of electrons in nanostructured materials such as thin films results in discrete quantum well states,[1,2] which have attracted intensive research interests in the view of both fundamental science and technological applications. Recent advances in growth technique have made it possible to control the thickness of metal thin films with atomic precision,[3] which provides the playground for modulating the physical properties of nanomaterials in a wide variety of aspects,[4−11] such as transport,[4] chemical reactivity,[5,6] growth[7] and magnetism.[8] The physical mechanism behind those modulations is the periodic occupation of quantum well states (QWSs), i.e., as the thickness of thin metal films increases, the quantum well states shift progressively in energy and periodically move unoccupied QWSs below the Fermi level. Thus many physical properties can be modulated in a controlled manner because of their close correlation with the electronic structure of the system. A prerequisite for those quantum controls relies on an efficient confinement of electron states, i.e., sharpness of QWSs. As the coherency of the reflection of electron states at the interface strongly affects the lifetime of QWSs,[1] it is crucial to modify the interface structure to increase the efficiency of electron confinement.

We have studied the effect of interface modification on QWSs of Pb ultrathin films by preparing Pb quantum islands on Si(111) substrates with both the pristine 7 × 7 surface and the modified R3 surface. Our scanning tunneling microscopy/spectroscopy (STM/STS) characterization shows that growing Pb films directly on the Si(111)-(7 × 7) surface generates many defects at the Pb/Si interface, which reduces the coherent reflection of electrons and locally broadens the QWSs of Pb thin films. Meanwhile, Pb films grown on the R3 surface have smoother interfaces and exhibit smaller variations in the linewidth of QWSs at different locations on the film surface. The QWSs of Pb films grown on the R3 surface are remarkably sharper than those on the 7 × 7 surface, which signifies the importance of interface modification in improving the electron confinement.

Experiments were conducted with a Unisoku low temperature scanning tunneling microscope. The Si(111) substrate (p-type with a resistivity of 0.2 Ω·cm) was cleaned using the standard procedure of current heating at a base pressure of 1×10^{-10} Torr for a few cycles until large scale defect-free 7 × 7 surface reconstruction was obtained. The Si(111)-Root3×Root3-Pb surface was prepared by depositing 3–4 monolayers (MLs) of Pb from a Knudsen cell followed by annealing to 300°C. For the Pb films on the 7 × 7 surface, Pb islands were prepared by depositing 7–8 MLs of Pb followed by annealing at room temperature for 30 min. For the Pb films on the R3 surface which is smoother, 7 ML of Pb was deposited followed by annealing at room temperature (RT) for 15 min to form Pb islands with comparable sizes to those grown on the 7 × 7 surface. The STS spectra were recorded using a lock-in technique with a bias modulation of 20 mV (rms value) at a frequency of 1991 Hz at a temperature of 77 K, while the tunneling gap was set at $V = 0.5$ V and $I = 0.1$ nA. An e-beam heated polycrystalline PtIr tip was used in the experiments, which is grounded for reference voltage.

The Pb islands grown on the Si(111)-7×7 substrate have wedge-shaped geometry, which spans across

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**Email: xcma@aphy.iphy.ac.cn

many underlying Si terraces while keeping their top atomically flat.[12] A Pb film of thickness between 7 ML to 30 ML can be routinely obtained in a single sample. Here the width of Si terraces is around 60 nm, which is far larger than the Fermi wavelength of Pb, electron confinement in the lateral directions can be safely neglected. For a certain thickness of Pb within an island, the confinement in the vertical direction is the same as that of a macroscopically uniform Pb film with the same thickness. Thus a single Pb island could contain Pb films of multiple thicknesses. Because the Fermi wavelength of Pb is 3.8 times the Pb lattice constant in the [111] direction, Pb islands are sufficient for studying quantum oscillation physics with many periods and even the beating mode on top of the finer oscillation.[5,9,13] As a result, the Pb island possessing the peculiar geometry provides an ideal system for investigating the quantum size effect with local probe techniques, which dramatically saves the effort of preparing macroscopic films for characterization with ensemble-average techniques.[4]

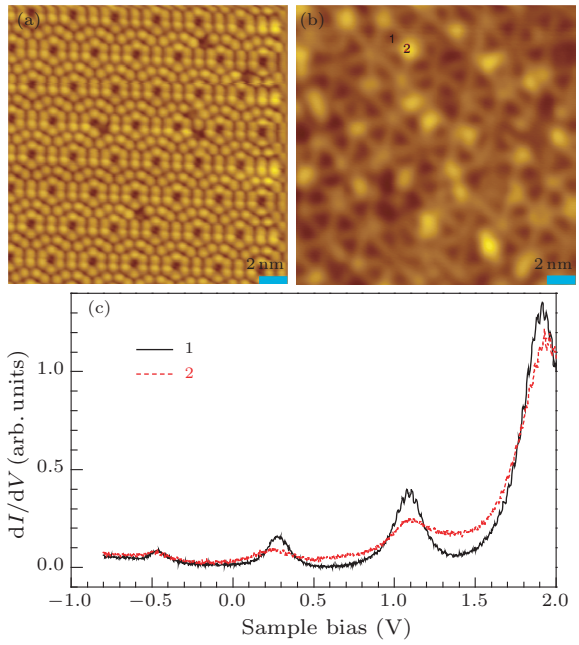

Fig. 1. Pb films on Si(111)-7 × 7 surface. (a) Atomic resolution image of Si(111)-7×7 surface ($V = 1.5$ V, $I = 1$ nA). (b) Typical STM image obtained on top of Pb film surface (film thickness: 14 ML) with 7 × 7 interface. Image condition is $V = 0.3$ V and $I = 1$ nA. Lattice structure of 7 × 7 reconstructions and superimposed defect spots can be clearly seen. (c) Differential conductance dI/dV spectra measured on two different locations (as indicated in (b)) of Pb film surface.

Pb islands grown directly on the Si(111)-7 × 7 surface generate many defects at the interface. As shown in Fig. 1(b), an STM image obtained on top of the Pb island exhibits the buried 7 × 7 interface reconstruction together with many defect spots. These defect spots are mainly located in the corner hole position of the pristine 7 × 7 reconstruction. Previous studies speculated that those defects could be generated by aggregation of Si adatoms from the pristine 7 × 7 reconstruction during growth of Pb and the remaining part of the 7 × 7 surface reconstruction is kept intact for reflecting the electronic states of Pb.[14] Tunneling spectroscopy measured at this area (presented in Fig. 1(b)) consists of series of peaks, which are ascribed to the quantum well states at the corresponding energy. Furthermore, dI/dV spectra show strong variations with different surface locations. As shown in Fig. 1 (b), point 2 is on the top surface of Pb islands but corresponds to a defect spot at the buried interface. The dI/dV curve obtained at point 2 (curve 2 in Fig. 1(b)) exhibits significantly broadened peaks compared to curve 1, which was obtained at point 1 and corresponds to the center of the buried 7 × 7 reconstruction. The interface defects result in incoherent scattering of the confined electron states and locally shorten the lifetime of quantum well states. Due to the high density of the buried defects, quantum well states of the Pb island surface heavily depend on the topographic locations. This makes the modulation of some locally related physical properties such as the Kondo effect of single molecular spin less well-defined.[8]

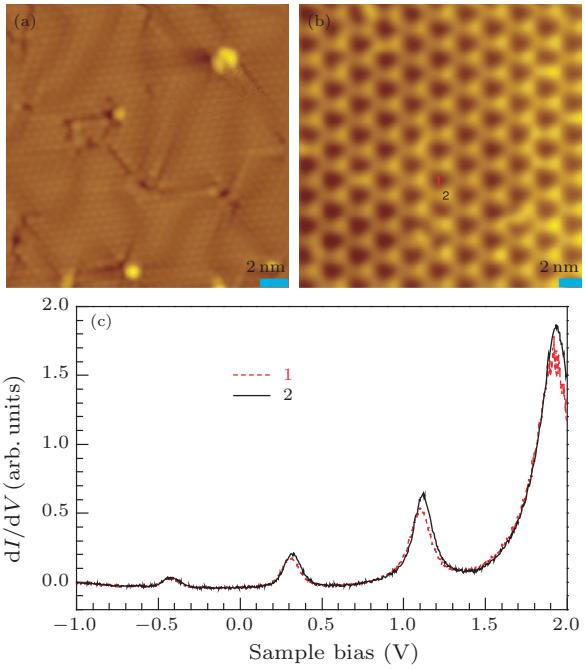

Fig. 2. Pb films on Si(111)-R3×R3 Pb surface. (a) Atomic resolution image of Si(111)-R3×R3 Pb surface with stripe incommensurate phase. ($V = 1.0$ V, $I = 1$ nA) (b) STM image of Pb film surface (film thickness: 14 ML) with R3 interface showing distinct hexagonal moiré pattern. ($V = 1$ V, $I = 0.1$ nA) (c) Tunneling spectra of Pb film obtained at two different surface locations of the moiré pattern labeled in (b).

Growth of a single ML thick Pb reconstruction on top of the Si(111) effectively removes the atomically

corrugated pristine 7×7 surface. Figure 2(a) shows the atomic resolution of the stripe incommensurate Si(111)-α R3 Pb reconstruction, which is a Pb-rich phase. There are several phases of Pb reconstruction on the Si(111) surface, depending on the content of Pb atoms per Si lattice.[15] Our studies show that regardless of the phases of Pb reconstruction at the first step in preparation, the wetting layer always shows the Pb-rich α phase after the growth of Pb islands. This demonstrates that the formation of Pb islands happens only after the formation of the α phase by feeding Pb atoms into the Pb reconstruction. The smoothness of the α phase can also be manifested by reduced the annealing time for growing Pb islands of comparable size as that on the 7×7 surface.

Figure 2(b) is an image obtained on top of a Pb island. Hexagonal moiré patterns at the interface due to stacking of Pb onto Si with different lattice constants can be clearly seen and the defects at the interface have been effectively removed. The lattice constant of the interface Pb atoms underneath the Pb islands has relaxed into its bulk value.[16] Tunneling spectra of Pb films show series of very sharp peaks. More importantly, the QWSs are well-defined with very small variations in the sharpness of QWS peaks at different locations relative to the moiré pattern. The small shift in energy of the QWSs at different surface locations arises from the contributions of different phase to the formation of QWSs at the interface,[16] which leads to a periodic modulation of the surface energy and has been utilized to grow nanocluster arrays on top of the Pb island.[17]

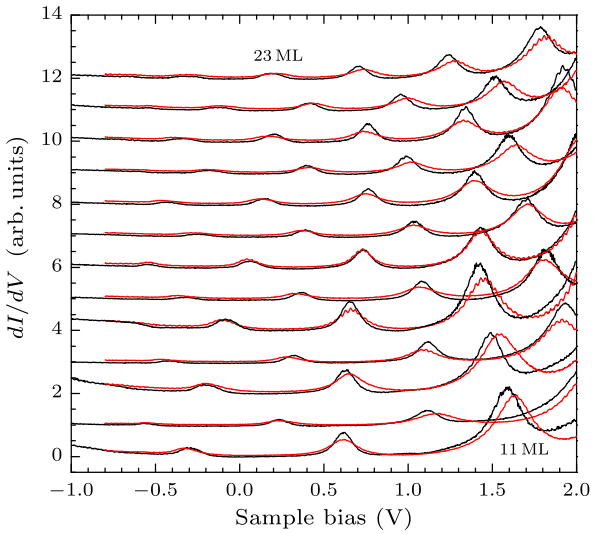

Fig. 3. QWSs of Pb films with different thicknesses and interfaces. Differential conduction (dI/dV) spectra measured on Pb films between 11 ML and 23 ML. Red curves and black curves were measured on Pb films with 7×7 and R3 interfaces respectively.

In order to compare QWSs of Pb films with the two different interfaces, we have measured two complete sets of QWS spectra with Pb thickness ranging from 11 ML to 23 ML. As is seen in Fig. 3, red curves are QWSs of Pb on the 7×7 surface and black curves are QWSs of Pb on the R3 surface. Energy positions of the two sets of QWSs coincide, which means that the absolute thicknesses of Pb for the two interfaces are the same. Because the R3 reconstruction is 1 atomic layer thick, the thickness of the noncrystalline wetting layer on the 7×7 surface is determined to be 1 ML. Peak widths of QWSs for the R3 interface are essentially smaller than those for the 7×7 interface, which demonstrates that the lifetime of the modified QWSs is enhanced. The lifetime of QWSs can be extracted by numerically fitting the QWSs with Lorentz peaks.[18] For example, the lifetimes of the lowest unoccupied QWS of 11 ML Pb on 7×7 and R3 interfaces are 195 meV and 125 meV, respectively. The lifetime of QWSs is composed of three terms,[1] i.e., electron-phonon scattering, electron-electron interaction and electron-defect scattering. The former two terms are related to the material properties and film thickness, which are the same for the Pb films under investigation. As a result, the difference in lifetime lies in the contributions from different electron-defect scattering at two interfaces. This is consistent with the degree of smoothness of the two interfaces. The interface modification not only removes local defects and make QWSs more well-defined, but could also be employed to improve the lifetime of QWSs.

In summary, modification of QWSs in Pb films has been performed by introducing a single atomic layer of R3-Pb reconstruction at the interface. Compared with the pristine 7×7 surface, R3-Pb reconstruction effectively removes the large density of subsurface defects that perturb the QWSs and enhances the lifetime of QWSs by reducing electron-defect scattering at the interface. The interfacial engineering[19] presented here provides a new approach for the study and control of quantum well states.[20]

References

[1] Chiang T C 2005 *Surf. Sci. Rep.* **39** 181
[2] Milun M, Pervan P and Woodruff D P 2002 *Rep. Prog. Phys.* **65** 99
[3] Jia J F, Li S C, Zhang Y F and Xue Q K 2007 *J. Phys. Soc. Jpn.* **76** 082001
[4] Guo Y, Zhang Y F, Bao X Y, Han T Z, Tang Z, Zhang L X, Zhu W G, Wang E G, Niu Q, Qiu Z Q, Jia J F and Xue Q K 2004 *Science* **306** 1915
[5] Ma X C, Jiang P, Qi Y, Jia J F, Yang Y, Duan W H, Li W X, Bao X H, Zhang S B and Xue Q K 2007 *Proc. Natl. Acad. Sci.* **104** 9204
[6] Jiang P, Wang L L, Ning Y X, Qi Y, Ma X C, Jia J F and Xue Q K 2009 *Chin. Phys. Lett.* **26** 016803
[7] Ma L Y, Tang L, Guan Z L, He K, An K, Ma X C, Jia J F and Xue Q K, Han Y, Huang S and Liu F 2006 *Phys. Rev. Lett.* **97** 266102
[8] Fu Y S, Ji S H, Chen X, Ma X C, Wu R, Wang C C, Duan

W H, Qiu X H, Sun B, Zhang P, Jia J F and Xue Q K 2007 *Phys. Rev. Lett.* **99** 226601
[9] Zhang Y F, Jia J F, Han T Z, Tang Z, Shen Q T, Guo Y and Xue Q K 2005 *Phys. Rev. Lett.* **95** 096802
[10] Qi Y, Ma X C, Jiang P, Ji S H, Fu Y S, Jia J F Xue Q K and Zhang S B 2007 *Appl. Phys. Lett.* **90** 013109
[11] Jiang P, Ma X C, Ning Y X, Song C L, Chen X, Jia J F and Xue Q K 2008 *J. Am. Chem. Soc.* **130** 7790
[12] Altfeder I B, Matveev K A and Chen D M 1997 *Phys. Rev. Lett.* **78** 2815
[13] Czoschke P, Hong H, Basile L and Chiang T C 2004 *Phys. Rev. Lett.* **93** 036103
[14] Altfeder I B and Chen D M 1998 *Phys. Rev. Lett.* **80** 4895
[15] Chan T L, Wang C Z, Hupalo M, Tringides M C, Lu Z Y and Ho K M 2003 *Phys. Rev.* B **68** 045410
[16] Altfeder I B, Narayanamurti V and Chen D M 2002 *Phys. Rev. Lett.* **88** 206801
[17] Lin H Y, Chiu Y P, Huang L W, Chen Y W, Fu T Y, Chang C S and Tsong T T 2005 *Phys. Rev. Lett.* **94** 136101
[18] Hong I P, Brun C, Patthey F, Sklyadneva I Y, Zubizarreta X, Heid R, Silkin V M, Echenique P M, Bohnen K P, Chulkov E V and Schneider W D 2009 *Phys. Rev.* B **80** 081409(R)
[19] Wu R, Wang L L, Zhang Y, Ma X C, Jia J F and Xue Q K 2010 *Chin. Phys. Lett.* **27** 026802
[20] Ricci D A, Miller T and Chiang T C 2005 *Phys. Rev. Lett.* **95** 266101