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Effects of atomic corrugations on electronic structures in $Pb_{1-x}Bi_x$ thin films

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We carried out experimental investigations of the geometric effect on the electronic behavior in $Pb_{1-x}Bi_x$ thin films by scanning tunneling microscopy and spectroscopy. Single crystal monolayer $Pb_{0.74}Bi_{0.26}$ and two-monolayer $Pb_{0.75}Bi_{0.25}$ $Pb_{1-x}Bi_x$ thin films were fabricated by molecular beam epitaxy, where large surface corrugations were observed. Combined with tunneling spectroscopic measurements, it is found that atomic corrugations can widely change the electronic behaviors. These findings show that the $Pb_{1-x}Bi_x$ system can be a promising platform to further explore geometry-decorated electronic behavior in two-dimensional metallic thin films.

Keywords: electronic contribution, geometric corrugation, scanning tunneling microscopy, Pb–Bi alloy films

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1. Introduction

Recent findings on the geometry-decorated electron correlation on two-dimensional (2D) systems show compelling electronic properties.[1,2] For example, in the Pb_3Bi/Ge (111) system, large Rashba splitting originating from strong spin–orbital coupling (SOC) elements Pb[3–5] and Bi[6–8] can be further enhanced by the asymmetric potential and the geometric phases originating from inversion symmetry breaking, which play important roles in inducing topological superconductivity.[1,9] In thin Pb films, substrates contribute to strengthening electron–phonon interactions by inducing out-plane electronic bonds,[10] and geometric corrugations induced by lattice distortion can modulate the quantum size effect (QSE)[11–13] that dominates the film properties. Moreover, intrinsic structural corrugations also exhibit pronounced metallic properties.[14] In a very recent work,[15] a new $3\sqrt{3}\times 3\sqrt{3}$ metallic phase was reported to show a large surface corrugation, which may have originated from the QSE[13,16,17] strain texture-dominated electronic behaviors[18,19] or from geometric symmetry breaking effects.[14]

In this study, we focus on low dimensional $Pb_{1-x}Bi_x$ alloy systems to explore the effects of the surface corrugations. In the 1-ML $Pb_{0.74}Bi_{0.26}$ and two-monolayer (2ML) $Pb_{0.75}Bi_{0.25}$ films prepared with the molecular beam epitaxy (MBE), surface corrugations as large as 0.15 nm have been observed by scanning tunneling microscope (STM). Strong electronic inhomogeneity directly corresponding to the surface corrugations was revealed in the conductance mapping by scanning tunneling spectroscopy (STS) measurements. Hence, the strong geometric effect can greatly decorate the electronic behaviors in a low dimensional $Pb_{1-x}Bi_x$ system.

2. Methods

Here we used n-type Si (0.001 Ω·cm–0.009 Ω·cm resistivity and a 0.1°–0.5° miscut angle toward [112]). First, a Si (111) wafer was degassed at 500 °C for 10 hours and then cleaned by flashing to ∼ 1000 °C for several cycles to obtain a Si (111)-7 × 7 reconstruction. One monolayer striped incommensurate Pb (SIC) substrates were prepared following the standard procedures,[20,21] in which Pb was deposited at 460 °C for around 5 min and then annealed at approximately 300 °C for around 2 min. Lead (purity 99.999%) and bismuth (99.999%) were evaporated with K-cell evaporators on SIC substrates at the flux rates of 0.35 ML/min and 0.17 ML/min (1 ML = 7.8×10^{14} atoms/cm^2), respectively. To obtain different x $Pb_{1-x}Bi_x$ thin films, we adjusted the flux rates of Pb and Bi sources by changing the source temperatures, and also changed Pb and Bi deposition times. 1-ML $Pb_{0.74}Bi_{0.26}$ films were prepared by depositing Bi on SIC for 3 min followed by 3 min of an annealing process at approximately 200 °C. 2-ML $Pb_{0.75}Bi_{0.25}$ films were prepared by co-depositing Pb and Bi atoms on SIC for 6 min followed by 6 min of a room-temperature (RT) annealing process. All samples were deposited in an MBE chamber at around 90 K. Then the prepared samples were transferred to the sample storage stage to perform the annealing processes. The rate of the increasing temperature was around 20 K/min–30 K/min for the temperature in the range of 90 K–200 K. It was estimated that the

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stage temperature became RT at around 1 hour. Experiments were performed on a commercial LT-STM system (Unisoku. 1500) with an RHK R9 controller. STM/STS measurements were carried out at 78 K with a constant current mode. STS measurements were obtained by using a lock-in amplifier with modulation amplitudes of 2 meV or 5 meV ($V_{\rm mod} = 2$ meV or 5 meV) and a lock-in frequency of 0.973 kHz.

3. Results

The growth of 1-ML $Pb_{1-x}Bi_x$ films has been performed in previous report.[15] In this work, it is stated that when $x = 0.26$, $3\sqrt{3} \times 3\sqrt{3}$, the PbBi metallic alloy phase can almost cover the whole film. We have performed surface corrugation studies on the 1-ML $3\sqrt{3} \times 3\sqrt{3}$ phase (marked as 1-ML $Pb_{0.74}Bi_{0.26}$ below), as shown in Figs. 1(a) and 1(b), which display the topography of the 1-ML $Pb_{0.74}Bi_{0.26}$. From the line profile results in Fig. 1(c), it is meaningful to conclude that the structure shows surface corrugation with a $\sim$ 0.15 nm amplitude. This type of large surface corrugation has also been found in 2-ML $Pb_{1-x}Bi_x$ thin films, which will be discussed later. In previous works, Pb or its alloy films showed small surface corrugations, and all of the atoms in one layer were almost sited around the same plane[9,10,13,15,20–25] and the electronic properties of the Pb thin films were determined by the electronic structure and geometric corrugations simultaneously.[11–13,25] Moreover, the layer plane deviation of the atoms has been studied in (Tl, Au)/Si (111)-7×7 films, where it was found out that such atomic corrugations could enhance low temperature metallicity,[14] and the electronic behavior could also be greatly modulated.[10,13,23,25] Here, we focus on the effects of the surface corrugations on the electronic structures in $Pb_{1-x}Bi_x$ thin films, and it is necessary to clarify whether the large surface corrugations originate from the electronic contribution or from the atomically geometric corrugations, or from both of them.

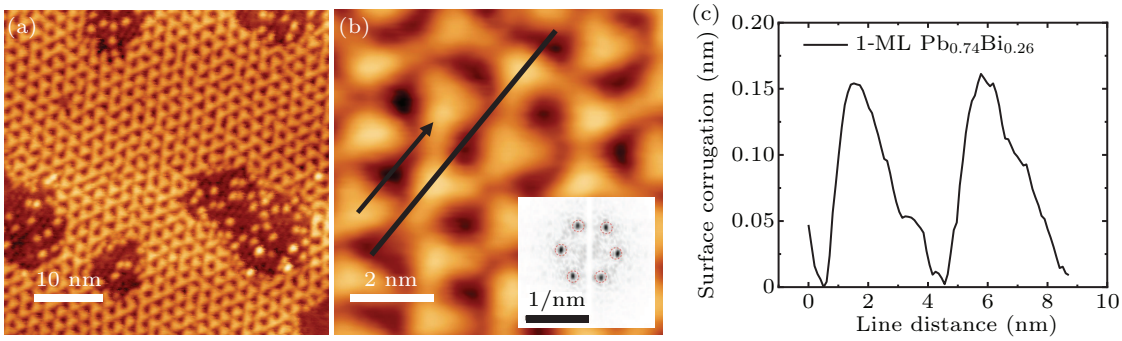


Fig. 1. (a) 60 nm×60 nm topography of 1-ML $Pb_{0.74}Bi_{0.26}$ films, $V = 1$ V, $I = 30$ pA. (b) 10 nm×10 nm topography of 1-ML $Pb_{0.74}Bi_{0.26}$ films, $V = 1$ V, $I = 50$ pA. The bottom-right inset image is the fast-Fourier transform (FFT) of the image. (c) Line profile of the black line in (b). The black arrow in panel (b) is the direction of the line profile.

First, we perform STS measurements to show how large surface corrugations affect the electronic properties in 1-ML $Pb_{0.74}Bi_{0.26}$. Figure 2(a) shows the dI/dV spectroscopy in the 1-ML $Pb_{0.74}Bi_{0.26}$. In contrast to the electronic states dominated simultaneously by interlayer and intralayer bonds between SIC and Si substrates,[1,10] surface corrugations also play important roles in determining electronic properties in 1-ML $Pb_{0.74}Bi_{0.26}$. Figure 2(a) shows a strongly modified energy peak between the bright region and the dark region around -0.4 eV, which means that a strong electronic inhomogeneity exists in the films. From the continuous measurements of the electronic states shown in Fig. 2(b), it is clear that the energy peak around -0.4 eV varies with the geometric corrugations, and the peak density is higher in the dark region and lower in the bright region. From Figs. 1(b) and 1(c), it can be recognized that the -0.4-eV energy peak corresponds to lower surface corrugation parts. Hence, it is concluded that due to large surface corrugations, the electronic states have been changed greatly, and the electronic properties in 1-ML $Pb_{0.74}Bi_{0.26}$ is closely related to the long-range ordered surface corrugations, 1-ML $Pb_{0.74}Bi_{0.26}$ is a strong geometry-decorated electronic system.

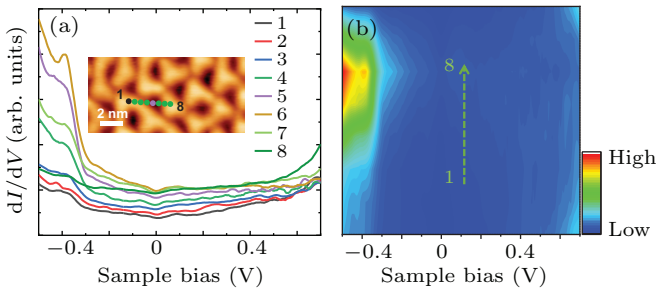


Fig. 2. (a) dI/dV line spectroscopy across eight points in 1-ML $Pb_{0.74}Bi_{0.26}$. (b) dI/dV density view from point 1 to point 8 in panel (a). The right color bar represents the density of the spectroscopy, and the upper colors represent higher densities.

To further verify that the surface corrugations are indeed atomically geometric corrugations rather than the electronic contribution in $Pb_{1-x}Bi_x$ thin films, we perform simultaneous STM topography and dI/dV conductance mapping measurements at different sample biases, as shown in Fig. 3. In thin films where the electronic behavior are determined simultaneously by electronic contribution and geometric corrugations,

the surface topography can be changed by changing the sample biases.[2,10–12] We also find these types of phenomena in the SIC region, as shown in Fig. 3, where the STM images consist of SIC in the left half and 1-ML $Pb_{0.74}Bi_{0.26}$ in the right half, both the topography and the dI/dV mappings in the SIC can be changed at different sample biases. While it is significant to demonstrate that 1-ML $Pb_{0.74}Bi_{0.26}$ shows no obvious topography transformation or modification at a wide sample bias range as shown in Fig. 3. Moreover, from the results of dI/dV conductance mappings, the evidence of the modification of electronic properties by the geometric effects is found. In 1-ML $Pb_{0.74}Bi_{0.26}$, as shown in Fig. 3, there is a clear contrast between the filled and empty electronic states in dI/dV mappings, and its empty states always show higher density of states in the bright regions, while its filled states always show higher density of states in the dark regions, a clear demonstration of the geometric effect. Similar to thin Tl–Au alloy films,[14] it is evident that the surface corrugations are atomically geometric corrugations in the 1-ML $Pb_{0.74}Bi_{0.26}$. Large atomic corrugations play important roles in determining the electronic behavior in thin $Pb_{1-x}Bi_x$ films and stabilizing the electronic states against sample bias changing at a wide energy range.

Furthermore, similar geometry-dominated electronic behavior is found in the 2-ML $Pb_{1-x}Bi_x$ films, such as the 2-ML $Pb_{0.75}Bi_{0.25}$ shown in Fig. 4. It is found that there is a 2-ML $Pb_{1-x}Bi_x$ phase, such as the bright spots in Figs. 4(a) and 4(b), which consists of an approximately 4.65-nm periodic phase in large scale and a $\sqrt{3}\times\sqrt{3}$ phase in atomic scale. This 2-ML $Pb_{1-x}Bi_x$ structure covers the whole substrate when x reaches 0.5, and we show the 2-ML $Pb_{0.75}Bi_{0.25}$ here to reveal its large surface corrugations. From continuous conductance measurements displayed in Fig. 4(c), it is found that the energy peak around −0.8 eV can be changed by the atomic corrugations in terms of density of states. Unchangeable topography are also confirmed at wide sample biases in the 2-ML $Pb_{0.75}Bi_{0.25}$, and strong geometry-decorated electronic behavior emerges in the 2-ML $Pb_{0.75}Bi_{0.25}$. Hence, it is evident that there are atomic corrugations in these types of highly ordered $Pb_{1-x}Bi_x$ thin films, which create large geometric corrugations and transform $Pb_{1-x}Bi_x$ thin films into geometry-dominated electronic systems.

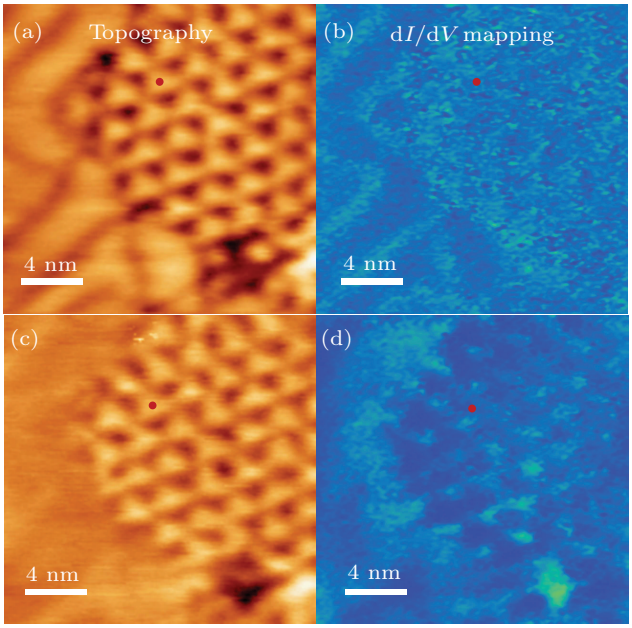


Fig. 3. 1-ML $Pb_{0.74}Bi_{0.26}$ topography and STS electronic conducting mappings for different sample biases V: (a) topography, $V = 1.5$ V; (b) dI/dV mapping corresponding to panel (a), $V = 1.5$ V; (c) topography, $V = -340$ mV; (b) dI/dV mapping corresponding to panel (c), $V = -340$ mV. For panels (a)–(d), $I = 50$ pA. The red spots represent the same position at different sample biases.

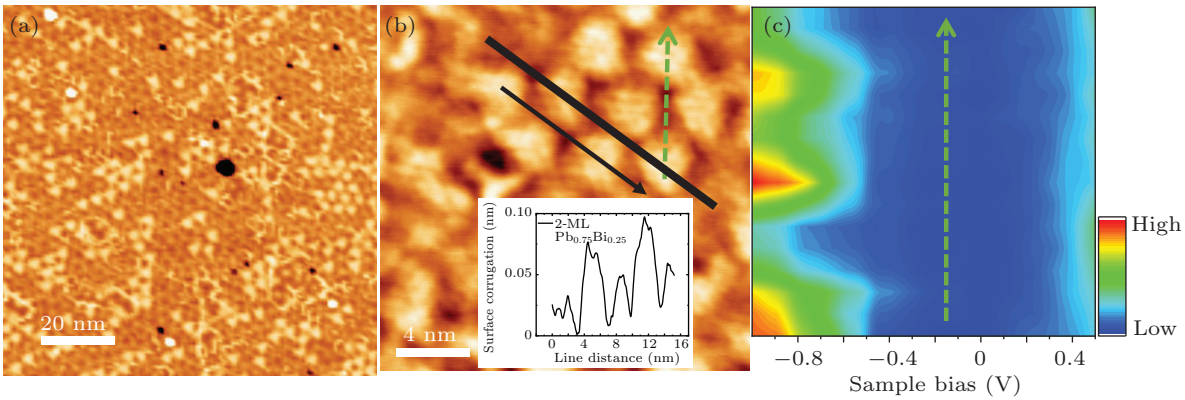


Fig. 4. 2-ML $Pb_{0.75}Bi_{0.25}$ STM images and STS: (a) 100 nm×100 nm topography of 2-ML $Pb_{0.75}Bi_{0.25}$ films, $V = 3.54$ V, $I = 50$ pA; (b) 10 nm×10 nm topography of 2-ML $Pb_{0.75}Bi_{0.25}$ films, $V = 0.24$ V, $I = 50$ pA. The bottom-right inset image is the line profile of the black line, and the black arrow is the direction of the line profile; (c) dI/dV density view in panel (b). The green line in panel (c) represents the exact STS measuring position along the green line in panel (b). The color bar on the right represents the density of the spectroscopy, and the upper colors represent higher densities.

4. Discussion

From the investigation of topography line profiles in 1-ML $Pb_{0.74}Bi_{0.26}$ and 2-ML $Pb_{0.75}Bi_{0.25}$, it is found that the films show ∼0.15 nm or ∼0.05 nm corrugations, respectively. By the analysis of conductance spectroscopy in $Pb_{1-x}Bi_x$ thin films, we confirm that there is electronic inhomogeneity across

the periodicity, which can be closely related to large atomic corrugations. For both of 1-ML $Pb_{1-x}Bi_x$ and 2-ML $Pb_{1-x}Bi_x$ thin films, Bi contents do not change the atomic corrugations in such large-corrugated Pb–Bi alloy phases. For lower Bi concentration x, less large-corrugated Pb–Bi alloy structures will be grown, while with higher Bi concentrations, more large-corrugated Pb–Bi alloy structures will be grown and even transformed the films to other alloyed structures.[15] By simultaneously performing STM topography and dI/dV conductance mappings in the films, the inhomogeneity of electronic states can be verified in the wide energy range and be closely determined by the atomic corrugations. These findings show that in highly ordered and corrugated $Pb_{1-x}Bi_x$ thin films there is a strong geometric effect originating from the large layer plane deviation atoms, and the electronic behaviors are strongly determined by the geometric corrugations.

5. Conclusion

In this study, by investigating the large surface corrugations with STM/STS in 1-ML $Pb_{0.74}Bi_{0.26}$ and 2-ML $Pb_{0.75}Bi_{0.25}$, it is evident that the surface atomic corrugations can widely change the electronic structures in thin $Pb_{1-x}Bi_x$ thin films. Our finds show that $Pb_{1-x}Bi_x$ thin films could be promising systems to unveil new metallic properties, such as inversion symmetry breaking with the geometric effect and enhanced metallicity in two-dimensional systems.

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