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Superstructures at Te/Au(111) interface evolving upon increasing Te coverage

Jiaqi Guan[a,b], Xiaochun Huang[a,b], Xiaofeng Xu[a,b], Shuyuan Zhang[a,b], Xun Jia[a,b], Xuetao Zhu[a], Weihua Wang[a,*], Jiandong Guo[a,b,c]

[a] Beijing National Laboratory for Condensed Matter Physics and Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China
[b] School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100049, China
[c] Collaborative Innovation Center of Quantum Matter, Beijing 100871, China

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ABSTRACT

By *in-situ* low temperature scanning tunneling microscopy, we systematically investigated the superstructure evolution at Te/Au(111) interface upon increasing Te coverage. Te atoms form one-dimensional $\sqrt{3}R30°$ chains at ~0.10 monolayer (ML) coverage. Two two-dimensional chiral superstructures, $(\sqrt{111}\times\sqrt{111})R4.7°$ and $(3\sqrt{21}\times3\sqrt{21})R10.9°$, are selectively formed with the Te coverage below and above 1/3 ML, respectively. The two chiral superstructures can be converted to each other reversibly by adding Te atoms or moderately annealing. A honeycomb-like superstructure, decorated with adatoms that are distributed in quasi-one-dimensional chains, is observed by further increasing the Te coverage to 4/9 ML. At the Te/Au(111) interface, an interfacial state at −0.65 eV to −0.55 eV below the Fermi level is also resolved by scanning tunneling spectroscopy. The formation of these Te-induced high-order superstructures is accompanied by relaxation of gold atoms in the surface layer, indicating a strong Te–Au interaction. Our work demonstrates a reliable method to fabricate Te nanostructures on noble metals in a controlled way.

1. Introduction

The adsorption of atoms on surfaces is not only the basis for further film growth and chemical reactions, but also reveals the interaction between adsorbates and substrate [1–6]. Gold surface is widely used as a template in atom and molecule adsorption [7–9], self-assembly [10,11], and surface-supported reactions [12,13]. On the close-packed Au(111) surface, which shows reconstructed herringbone patterns due to the formation of mesoscopic stress domains [14,15], the investigation of adsorption behaviors provides intriguing physical insights into the adsorbate-substrate interaction. Previous reports have shown that the adsorption of oxygen, sulfur, chlorine, tellurium, gadolinium atoms as well as trimethylphosphine and alkanethiolate molecules can lift the herringbone reconstruction by introducing compressive stress [5,16–24], while alkali metal atoms can enhance the corrugation of topmost Au layer [6]. Additionally, the surface states of Au(111) are also modified due to the adsorbate-substrate interaction [25–28].

Tellurium is the heaviest non-radioactive element in chalcogen group. The bulk Te crystal is a mid-infrared semiconductor with a band gap of 0.33 eV [29], and its band gap increases monotonically with decreasing thickness, reaching 0.92 eV in monolayer Te film [30]. The adsorption of Te atoms on Au(111) has been investigated by various experimental methods, including low-energy electron diffraction (LEED), electrochemical-scanning tunneling microscopy (EC-STM), EC-atomic force microscopy (EC-AFM) and Auger electron spectroscopy (AES) [33–39]. It is revealed that the Te adsorption structures evolve with Te coverage, but discrepancies exist in the assignment of the Te adsorption superstructures. Previous LEED study by Nagashima et al. suggested non-commensurate $(\sqrt{3}\times\sqrt{3})R30°$ patterns at a nominal Te thickness of 0.15 nm [32], and (12×12) and (13×13) structures are proposed by Stickney et al. to describe the Te structure at a coverage of ~1/3 ML based on EC-STM experiments [21,34]. However, the EC-AFM studies by Ikemiya et al. reported a $(\sqrt{3}\times\sqrt{3})R30°$ structure without larger periodicity [36], and Hayden et al. reported a (12×12) structure at a Te coverage of 0.42 ML [35]. In the earlier EC-STM and EC-AFM studies of Te layers on Au(111), the adsorption structures were inevitably influenced by the solution, and the investigations on Te superstructure were limited by the image quality. To reveal the superstructure evolution at Te/Au(111) interface upon Te coverage, it is necessary to carry out the experiments in ultra-high vacuum (UHV) condition with Te coverage controlled in a well-defined way. Schouteden and co-workers studied the adsorption of residual Te atoms on Au(111), and revealed the strong interaction between Te adatoms and Au(111) surface, leading to scattering and confinement of the Au surface state near the Fermi level [24]. However, a systematic study of Te adsorption on Au(111) surface with

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[*] Corresponding author.
E-mail addresses: weihuawang@iphy.ac.cn (W. Wang), jdguo@iphy.ac.cn (J. Guo).

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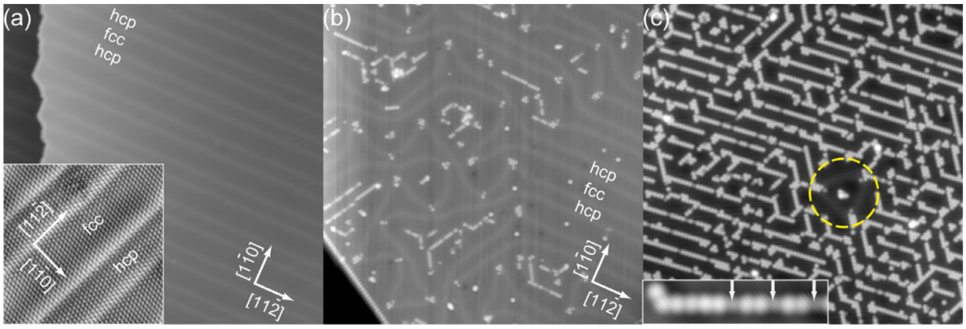

Fig. 1. (a) STM image (50×50 nm^2, 3.0 V/50 pA) of clean Au(111). The inset shows the atom-resolved image (1.0 V/200 pA). (b) and (c) STM images of Te atoms on Au(111) at coverage of 0.02 ML (50×50 nm^2, 1.2 V/70 pA) and 0.10 ML (40×40 nm^2, 1.0 V/50 pA), respectively. The inset shows the magnified STM image (6×1.4 nm^2, 0.2 V/50 pA) of Te chain on Au(111) with white arrows indicating the dim Te atoms.

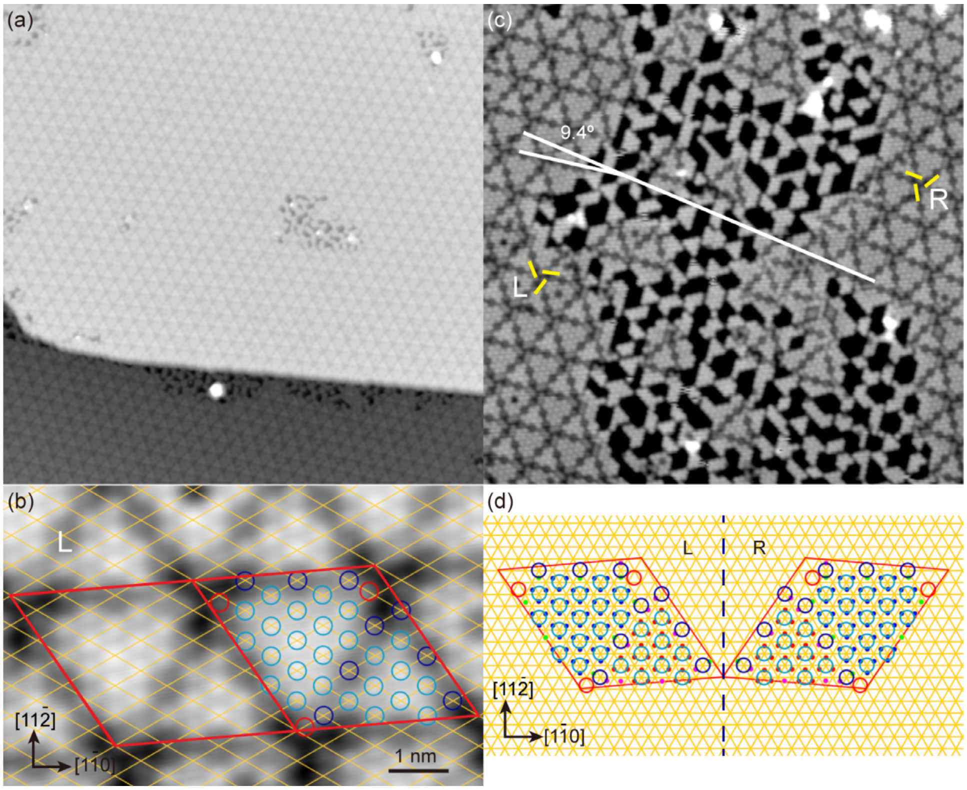

Fig. 2. (a) STM image (100×100 nm^2, 2.0 V/50 pA) of Te atoms on Au(111) at coverage of 0.24 ML. (b) Zoom-in STM image (8×5 nm^2, −0.025 V/50 pA) of the rhombus phase. The $(\sqrt{3} \times \sqrt{3})$ lattice of Au(111) are indicated by the yellow grids, and the $(\sqrt{111} \times \sqrt{111})R4.7°$ unit cells are shown by the red frames. (c) STM image (40×40 nm^2, 1.5 V/100 pA) showing two types of domains coexist with each other. The left and right chiralities are illustrated by the yellow lines. (d) A possible adsorption model of the $(\sqrt{111} \times \sqrt{111})R4.7°$ rhombus phase. The bright Te atoms are represented by cyan circles, the dim Te atoms by violet and red circles in (b) and (d). The first layer gold atoms in *fcc*- and *hcp*-positions with respect to the second layer are represented by blue and red dots (or red and blue dots), and the second layer gold atoms are represented by grids. The pink and green dots represent the *fcc*- and *hcp*-gold atoms at the edge of *fcc*- and *hcp*-domains. The third layer gold atoms are not plotted. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

different Te coverages is still lacking. Moreover, Te monolayer was recently used as a buffer layer to decouple blue phosphorus film from Au substrate [31], which raises a question about the interfacial state at the Te/Au interface. However, this interfacial state has never been addressed by experiments.

Herein by precisely controlling the Te coverage via molecular beam epitaxy (MBE), we obtain a series of superstructures at the Te/Au(111) interface with large periodicities and systematically investigate their evolution upon Te coverage by *in-situ* low-temperature STM and scanning tunneling spectroscopy (STS). Te atoms form one-dimensional (1D) chains at low coverage, and two-dimensional (2D) superstructures at higher coverage, including two chiral structures near 1/3 monolayer (ML) and a honeycomb-like structure at ~4/9 ML. The formation of these superstructures is accompanied by the relaxation of Au(111) reconstructed lattice. Moreover, we detect an interfacial state of the Te/Au(111) system, which manifests the interaction between Te and Au substrate.