

Supporting Information for

Two-Dimensional Au Lattices Featuring Unique Carrier Transport Preference and Wide Forbidden Gap

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Table of contents

- S1. The evolution of RHEED images for different Au structures on Si surface.
- S2. First-principles calculations for different Au structures on Si surface.
- S3. Band dispersion for calculating the effective mass of electrons and holes.

S1.

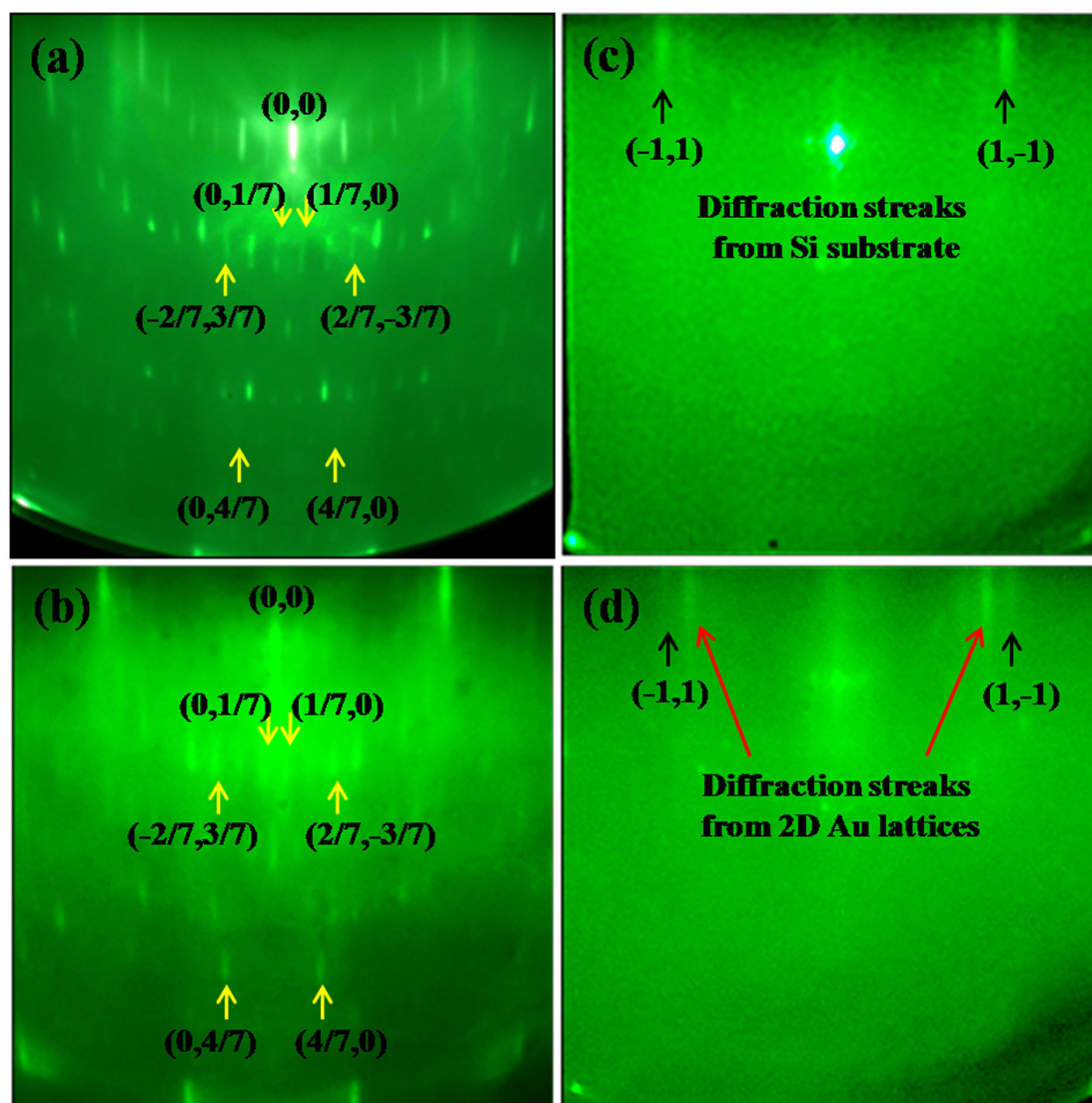


Figure S1 | RHEED pattern of (a) clean Si(111)-7 \times 7 surface, and 7 \times 7 surface with (b) Au_6Si_3 clusters, (c) 1L Au clusters, and (d) 2D Au lattices. The yellow arrows with numerical representation in (b) mark the specific positions of the diffraction spots which are enhanced after the formation of Au_6Si_3 clusters. The black arrows in (c) mark the diffraction spots (-1,1) and (1,-1) from the Si substrate. While the red arrows in (d) indicate two new diffraction streaks appeared after the formation of 2D Au lattices.

S2.

In order to deduce the possible configuration of 2D Au lattices, the adsorption

energies of the 1L and 2L Au atoms were calculated by the first-principles simulations within the framework of generalized gradient approximation, using the plane-wave density functional theory Vienna *Ab Initio* Simulation Package. The models were periodically repeated by an atomic slab consisting of four Si layers (200 Si atoms) and two Au₆Si₃ clusters on each HUC (the atomic configuration is referenced to our previous work,¹ with *n* additional Au atoms adsorbed on the top and 49 H atoms bonded at the bottom. A 12 Å thick vacuum layer was applied for separating the slab to form a surface. Ionic potentials were described by ultrasoft pseudopotentials.

According to the symmetry of Au₆Si₃-Si(111) surface, three possible adsorption sites were considered for an additional Au atom, as showed in Fig. S2(a). To compare the stability among these configurations, the adsorption energies were calculated by using the following formula:

$$E_{ads} = E_{Au+Au_6Si_3/Si7\times7} - [E_{Au_6Si_3/Si7\times7} + E_{Au}],$$

where $E_{Au_6Si_3/Si7\times7}$ and E_{Au} are total energies of the unit cell of Au₆Si₃-Si(111) surface and a single Au atom, respectively. The results listed in Table S1 suggest that, the Au atom on H₃ site has an adsorption energy of -3.83 eV, which is respectively 0.70 and 0.09 eV lower than that of AT and D site. It suggests that, after the formation of Au₆Si₃ cluster, the additional Au atom has preference to occupy the H₃ site, which is consistent with our STM observations that the three Si corner adatoms (Fig. 2(b2)) are invisible after the Au deposition increased to 0.4 ML (1L Au-Si layer, Fig. 2(c2)). Correspondingly, the RHEED pattern from Si(111)-7×7 is further weakened (Fig. 2(c1)) compared with that of the Au₆Si₃-Si(111) surface. Therefore, the most probable atomic structure of 1L Au-Si surface is deduced in Fig. S2(b). Based on this model, the atomic structure of 2L Au layer (2D Au lattices) was further studied, in which two possible adsorption sites were considered for an additional Au atom, as showed in Fig. S2(b). Similarly, the adsorption energy is expressed as:

$$E_{ads} = E_{Au+1L/Si7\times7} - [E_{1L/Si7\times7} + E_{Au}],$$

where $E_{1L/Si7\times7}$ is total energy of the unit cell of 1L Au-Si surface. The calculation results listed in Table S2 suggest that the adsorption energy of CT site is -3.81 eV, about 0.06 eV lower than that of the D site (-3.75 eV). Such energy difference means a preference for the 2L Au atoms locating on top of 1L Au-Si cluster rather than on top of the dimer site. These theoretical results combining with the RHEED and STM observations further serve as favorable evidences for the speculation that the Kagome-like lattices observed at +2.5 V may not actually reflect the geometric morphology of the surface, but likely originate from the electronic coupling effect in 2D Au lattices.

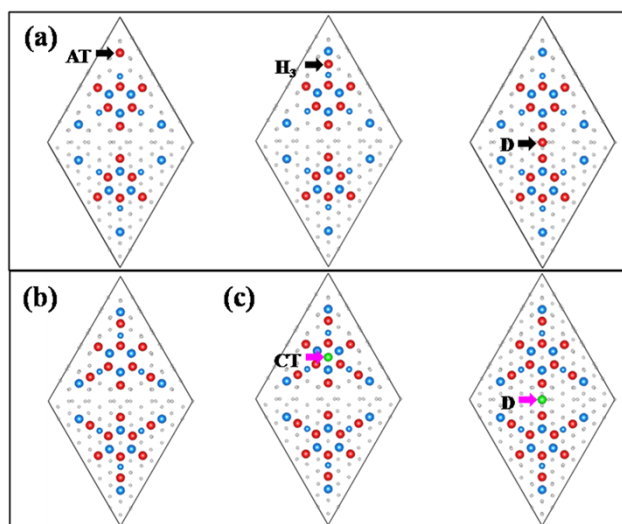


Figure S2 | (a) Top view of the structural models of Au_6Si_3 -Si(111) surface with one additional Au atom located on three possible adsorption sites (as indicated by the black arrows): AT site (top of Si corner adatom of Si(111)- 7×7 surface), H_3 site (hexagonal threefold coordinated position between Au_6Si_3 cluster and Si corner adatom), and D site (top of dimer site). (b) Top view of the most stable structural model of 1L Au-Si surface. (c) Top view of the structural models of 1L Au-Si surface with one additional Au atom located on two possible adsorption sites (as denoted by the purple arrows): CT site (center top of Au_6Si_3 cluster), and D site. The small blue, large blue, red, and green balls represent the Si rest atoms, Si adatoms, 1L Au atoms,

and 2L Au atoms, respectively.

Table S1 the adsorption energies for one Au atom at three possible adsorption sites on Au₆Si₃-Si(111) surface.

Adsorption site	AT	H ₃	D
Adsorption energy (eV)	-3.13	-3.83	-3.74

Table S2 the adsorption energies for one Au atom at two possible adsorption sites on 1L Au-Si surface.

Adsorption site	CT	D
Adsorption energy (eV)	-3.81	-3.75

S3.

The band dispersion of 2L Au structure on Si(111)-7×7 unit cell was obtained by the first principles simulations within the framework of generalized gradient approximation, using the plane-wave density functional theory Vienna *Ab Initio* Simulation Package. The effective mass of electrons and holes were approximately calculated from the band dispersion at Γ point (the blue arrow marked the conduction band 1 for electrons, and the red arrow marked the valence band 2 for holes). As can be seen from the band dispersion, the energy gap, without considering the carrier transport behavior, should be really small, which demonstrates that the measured large energy gap for continuous 2D Au lattices not only originate from the LDOS of 2L Au structure on each Si(111)-7×7 unit cell, but also modulated by the in-plane carrier transport preference after the formation of the continuous 2D lattices.

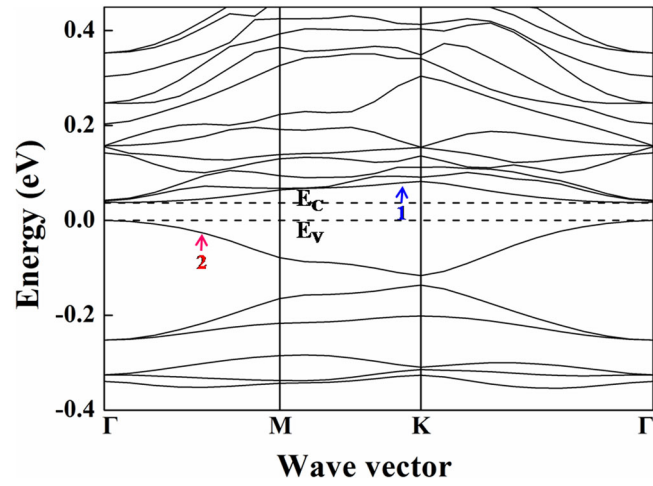


Figure S3 | Band dispersion of 2L Au structure on Si(111)-7×7 unit cell. The conduction and valence bands marked by the blue and red arrows were used for calculating the effective mass of electrons and holes, respectively. E_C and E_V represent the bottom of conduction band and the top of valence band, respectively.

1. Y. Wu, Y. Zhou, C. Zhou, H. Zhan, and J. Kang, *J. Chem. Phys.*, 2010, **133**, 124706.