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Unconventional electronic structure of silicene on Ag/H-Si(111)

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ABSTRACT

We fabricated monolayer silicene on a thin silver (Ag) crystal film grown on hydrogen-terminated Si(111), and studied the crystal and the electronic structure by low-energy electron diffraction (LEED) and high-resolution angle-resolved photoemission spectroscopy (ARPES). The fabricated sample exhibits the 3×3 LEED pattern characteristic of silicene and the band dispersions with the periodicity of the silicene Brillouin zone, indicative of successful fabrication of silicene on Ag/H-Si(111). However, we observed no signature for the Dirac-cone state at the K point, while the observed band structure exhibits an unexpected van-Hove singularity with a saddle point at the binding energy of 1 eV. Such a singularity has not been reproduced in so-far existing band structure calculations, requesting sophisticated calculations incorporating the complicated strong hybridization between silicene and Ag orbitals.

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1. Introduction

Silicene, a two-dimensional honeycomb sheet of silicon atoms like graphene, has attracted much attention since it is expected to show novel quantum phenomena such as the gate-voltage-induced topological phase transition and the quantum spin Hall effect [1–3]. It is theoretically predicted that free-standing monolayer silicene has a nearly massless Dirac-cone state composed of the Si $3p$ π and π^* bands with a small band gap due to the spin-orbit interaction [1] and the strong buckling of Si atoms [4]. The nearly massless nature of π π^* bands in silicene has been verified both experimentally and theoretically in bulk crystal of calcium-intercalated multilayer silicene, CaSi_2 [5,6]. On the other hand, it has been reported that monolayer silicene grown on a substrate [7–13] exhibits an electronic structure markedly different from that expected for free-standing silicene. Further, the experimental results themselves are not necessarily consistent with each other. For example, angle-resolved photoemission spectroscopy (ARPES) study on monolayer silicene grown on a Ag(111) single crystal [8,10–12] reported a massive Dirac-cone state with a sizable (~ 0.3 eV) band gap at the K point in the Brillouin zone (BZ) [8], which is two orders of magnitude larger than the spin-orbit gap predicted from the theory [1]. Other ARPES studies reported that the band structure shows an unusual

saddle point at the M point [10,11] or a massless Dirac-cone-like dispersion midway between the M and K points [12]. These experimental results strongly suggest that the electronic structure of silicene on a Ag substrate is much more complicated than that expected from the theoretical calculation for free-standing silicene, probably due to the strong interference from the substrate. It is thus quite important to study the substrate effect on the electronic structure of silicene, not only for better understanding of the intrinsic electronic properties of genuine silicene itself but also for its incorporation into electronic devices where silicene would be integrated on a substrate.

In this paper, we report a high-resolution ARPES study on monolayer silicene fabricated on a Ag(111) thin film grown on a hydrogen-terminated semiconducting Si(111) substrate. Our ARPES results have revealed the existence of σ bands dispersing along the K-M cut in the 1×1 silicene Brillouin zone (BZ), indicating the formation of stable sp^2 hybridized orbitals originating in silicene. Intriguingly, the Dirac-cone state near the Fermi level (E_F) expected in free-standing silicene is absent in the ARPES-derived band dispersions, whereas an unexpected van-Hove singularity was observed around the K point. We discuss implications of these ARPES results in comparison with previous ARPES reports of silicene on Ag(111) single crystal.

2. Sample fabrication and experiments

A thin Ag(111) single crystal film (thickness: ~ 20 monolayer), on which silicene was fabricated, was prepared by depositing Ag

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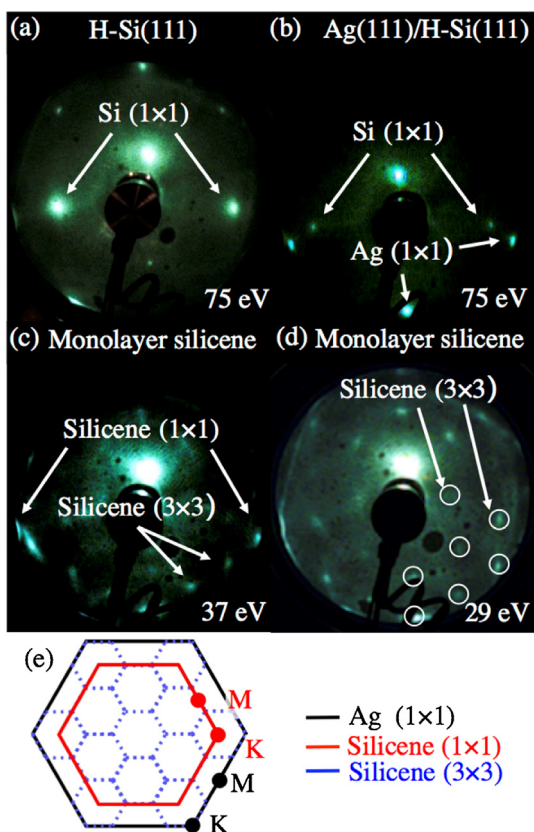


Fig. 1. (Color online): LEED patterns of (a) H-terminated Si(111), (b) Ag(111) thin film on H-Si(111), and (c, d) monolayer silicene on Ag(111)/H-Si(111), measured with the primary electron energy of 75, 75, 37 and 29 eV, respectively. (e) Schematic view of the Brillouin zones of Ag(111) (black solid line), 1×1 monolayer silicene (red solid line), and 3×3 monolayer silicene (blue dashed line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.).

atoms on a hydrogen-terminated *n*-type Si(111) crystal [14–17]. Fig. 1(a) and (b) shows the low-energy-electron-diffraction (LEED) patterns of the hydrogen-terminated Si sample before and after Ag deposition, respectively. Monolayer silicene was fabricated by subsequent deposition of Si atoms on the Ag/H-Si(111) substrate at 270 °C under a vacuum of 5×10^{-10} Torr. As shown in Fig. 1(c) and (d), the LEED pattern clearly exhibits the 3×3 and 1×1 spots, in good agreement with that reported for monolayer silicene on Ag(111) single crystal [7–12]. It is noted here that keeping the substrate at this high temperature (270 °C) during Si deposition is essential for fabricating silicene on Ag(111) and the hydrogen-termination of Si(111) substrate is also important to keep the Ag(111) crystal structure even at this high temperature. This is supported by the experimental fact that a Ag film grown without hydrogen-termination of Si(111) substrate shows a weak $\sqrt{3} \times \sqrt{3}$ pattern at ~ 200 °C owing to the diffusion of Ag atoms [17], unlike the hydrogen-terminated counterpart showing the 1×1 pattern. After thus confirming that monolayer silicene was successfully fabricated on Ag/H-Si(111), we transferred the sample to the ARPES chamber without exposing the sample to air. ARPES measurements were carried out using a Scienta-Omicron SES-2002 spectrometer with a high-flux He discharge lamp and a toroidal grating monochromator. The He I α (21.218 eV) line was used to excite photoelectrons. The energy and angular resolutions were set at 16 meV and 0.2° respectively. The sample was kept at 30 K during measurements. The Fermi level (E_F) was referenced to that of a gold film deposited onto the sample holder.

3. Results and discussion

Fig. 2(a) shows the valence-band ARPES spectra of Ag(111)/H-Si(111) (before Si deposition) measured along the Γ KM cut in the Ag(111) BZ. To see more clearly the dispersive features, we show in Fig. 2(b) the experimental band structure obtained by plotting the second-derivative intensity of ARPES spectra as a function of wave vector (k_x) and binding energy. In Fig. 2(b), we observe two categories of bands for the Ag(111) film; one is a bundle of several less dispersive bands located at 4–6.5 eV, and another is a highly dispersive single band in the energy range from 4 eV to E_F and apparently crosses E_F midway between the Γ and K points. The bundle of less dispersive bands is attributed to the Ag 4*d* orbitals, while the steep single band which crosses E_F is assigned to the Ag *sp* hybridized band which forms a large free-electron-like Fermi surface [10,11,18,19].

Fig. 2(c) and (d) displays the ARPES spectra and the corresponding band dispersions for silicene/Ag(111)/H-Si(111), respectively. One immediately finds that the Ag 4*d* bands located at 4–6.5 eV show a negligible change even after silicene formation on Ag(111)/H-Si(111), suggesting that the Ag 4*d* orbitals are not strongly influenced by the formation of silicene. Even when there exists a small hybridization, it may be smeared out by the ~ 50 times larger photo-ionization cross-section of Ag 4*d* orbitals than that of Si 3*s*/3*p* orbitals [20]. In contrast, as highlighted by a dashed curve in Fig. 2(c), a new band emerges at 2.7–4.0 eV at around the K point of the Ag BZ in silicene/Ag(111)/H-Si(111). Judging from the good agreement in the dispersive feature with the calculations for free-standing monolayer silicene [21,22], this newly emerging band is assigned to the σ band originating from the Si *sp*² hybridized orbital, consistent with previous ARPES results of silicene on Ag(111) single crystal [10,11]. The 3×3 periodicity of σ band is not clearly seen in the experiment, suggesting that the 3×3 periodic potential is relatively weak compared to that of the 1×1 structure. Another remarkable feature in the band structure of silicene/Ag(111)/H-Si(111) is the presence of a Ag *sp*-like band at around the K point in the silicene BZ, as visualized in Fig. 2(d). Intriguingly, this band does not cross E_F along the Γ KM cut unlike the Ag *sp* band in the Ag(111) film [see Fig. 2(b)] and instead appears to fold back at 1 eV at the K point of the silicene BZ. This behavior of band dispersion looks to resemble the massive Dirac-cone-like state reported for monolayer silicene on Ag(111) [8]. However, as shown in the following, this band shows an anomalous band dispersion which is hardly explained in terms of the massive Dirac-cone band.

Fig. 3(a)–(d) shows the plots of second-derivative ARPES intensity near E_F along four representative *k* cuts around the K point [cuts A–D in Fig. 3(e)], together with the peak position obtained by the numerical fitting of the energy distribution curves. Note that cut A is perpendicular to other three cuts (B–D). Now one can see more clearly that the band dispersion for cut A (Γ KM cut) exhibits a band folding at the K point. On the other hand, the band dispersions along cuts B–D commonly display an electron-like dispersion with the bottom at $k_y = 0 \text{ \AA}^{-1}$. This experimental fact that a single band exhibits a holelike dispersion along cut A and at the same time an electronlike dispersion along a cut perpendicular to it obviously indicates that this band forms a van-Hove singularity (vHs) with a saddle point at the K point as illustrated in Fig. 3(f).

Now we compare the present ARPES result with that of previous study of monolayer silicene on Ag(111) single crystal as well as with the band calculations. The previous ARPES study [8] observed a holelike dispersion along the Γ KM cut in qualitatively good agreement with the present observation, proposing that there is a massive Dirac cone at the K point in silicene on Ag(111). However, the previous study lacked the measurement perpendicular to the Γ KM cut to confirm whether the band dispersion really has a Dirac-cone shape. In the present study, on the other hand, we

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