

Orbital character of valence electronic states of epitaxial silicene on $\text{ZrB}_2(0001)$ thin film surfaces

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Introduction

While free-standing silicene is yet hypothetical, two-dimensional Si honeycomb lattices have recently been prepared on several metallic substrates such as Ag(111) [1] and $\text{ZrB}_2(0001)$ [2]. The structural and electronic properties of epitaxial silicene phases are, however, expected to be significantly altered from those of free-standing silicene. In order to resolve this complex matter for silicene on $\text{ZrB}_2(0001)$, new angle-resolved photoelectron spectroscopy (ARPES) data have been interpreted using a modified approach in density functional theory (DFT) first-principles calculations.

Experimental and Computational

ARPES spectra of epitaxial silicene on $\text{ZrB}_2(0001)$ thin films prepared on Si wafers [2] have been obtained with the SCIENTA SES200 analyzer at BL13B using the photon energy of 43 eV. First-principles calculations have been performed within the DFT and a generalized gradient approximation (GGA). A new conceptual unit cell that contains *only a single* Si atom for silicene, has been introduced in order to better represent the spectral weight as expressed by the imaginary part of the one-particle Green's function.

Results

ARPES spectra obtained along the $\bar{\Gamma} - \text{K}_{\text{Si}}$ direction and in a wide energy range, are shown in Fig. 1. The spectra are similar to those reported previously for the low-binding energy region [2,3]. The comparison of the ARPES spectra with the band structure of the so-called “planar-like” phase [4] at a lattice constant $a = 6.480 \text{ \AA}$ of the $(\sqrt{3} \times \sqrt{3})$ unit cell of silicene shows an astonishing and excellent quantitative agreement in terms of the energies and dispersions of electronic states.

All silicene-derived bands are hybridized to some extent with Zr d electronic states, which is consistent with non-negligible interactions at the interface. In particular, while the upward curved bands X_2 and X_3 in the vicinity of the $\text{K}_{\text{Si}}(1 \times 1)$ point have major contributions from Si p_z orbitals, at the same time, they hybridize with the Si s , p_x , p_y and the Zr d orbitals. As already suggested in our previous work [2], these states are therefore of partial π character. Bands X_6 and X_7 also have strong contributions of p_z orbitals and can be classified therefore as π bands.

On the other hand, bands S_1 and S_2 have almost sole contributions from d orbitals of the outermost Zr layer. This confirms that these states are diboride surface states [2,3] indeed that survive upon the formation of silicene.

The results resolve not only the orbital character of the hybrid silicene-diboride interface but also the previously controversial issue related to the structural configuration of silicene on the $\text{ZrB}_2(0001)$ surface that is now considered to be close to the “planar-like” model structure.

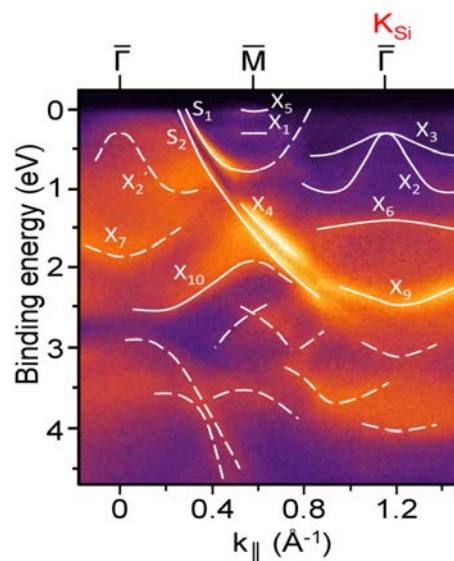


Fig. 1: ARPES spectra of epitaxial silicene on the $\text{ZrB}_2(0001)$ surface along the $\bar{\Gamma} - \text{K}_{\text{Si}}$ direction.

References

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