Engineering Bismuthene via Confinement Heteroepitaxy: Hydrogen-Controlled Structure and Spin Texture

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Confinement heteroepitaxy, which describes the intercalation of species beneath epitaxial graphene on SiC, has emerged as a versatile approach for synthesizing environmentally protected two-dimensional materials at well-defined interfaces [1]. Recently, considerable attention focused on Bi-intercalation of the graphene zero layer because it was reported to form a ($\sqrt{3}\times\sqrt{3}$) reconstructed β phase [2,3] that can be reversibly transformed – by hydrogenation or dehydrogenation – into the large-gap quantum spin Hall insulator bismuthene [4,5].

Using a combination of normal incidence X-ray standing wave imaging, spin- and angle-resolved photoelectron spectroscopy, and density functional theory, we demonstrate an H-induced change of the Bi adsorption site, which is accompanied by a contraction of the Bi layer. The atoms of the β phase occupy T_4 hollow sites with 1/3 monolayer coverage – a configuration that prohibits bismuthene formation. Partial H-passivation of the SiC interface contracts the lattice to a honeycomb structure with 2/3 monolayer coverage and shifts the atoms to the T_1 on top position. This allows for Bi-Bi hybridization, which, together with the remaining Bi-Si bond, establishes the orbital composition around the Fermi level required for the formation of bismuthene's Dirac-like band structure (see Fig. 1 (a)).

Due to the covalent bonding to the substrate, both Bi-rich structures are prone to strong mirror symmetry breaking, which leads to Rashba splitting of the valence bands. For the β and bismuthene phases, we demonstrate the lifting of the spin degeneracy, as well as the expected Rashba-type spin-momentum locking. Figs. 1 (b) and (d) show the in-plane polarizations of an exemplary energy distribution curve (EDC) that cuts through the bismuthene valence bands close to the K point (see black rectangle in (a)). In both directions, we observe significant polarization of the photocurrent that changes sign along the energy axis. This gives rise to two oppositely polarized components of the EDC, as shown in (c) and (e), which are attributed to bismuthene's spin-split valence bands.

References:
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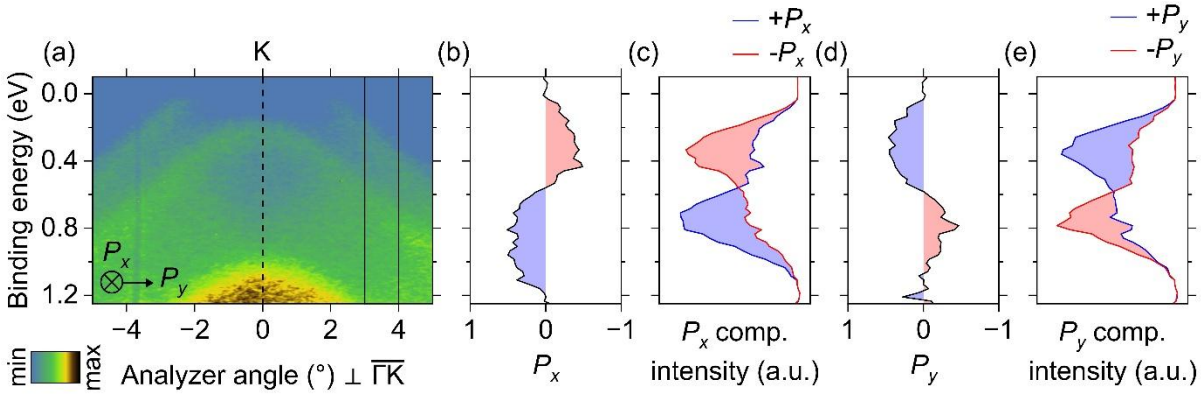

Fig. 1: (a) Energy-angle map of bismuthene at K, perpendicular to the $\overline{\Gamma K}$ direction. The black rectangle indicates the region from which the spin-resolved EDC was extracted. (b) and (d) show the respective polarizations in the two in-plane directions: $P_x \parallel \overline{\Gamma K}$, and $P_y \perp \overline{\Gamma K}$. (c) and (e) display the corresponding EDC component intensities. Red and blue colors denote negative and positive polarization, respectively.