## **Supporting Information**

## Layer-controlled evolution of electron state in the silicene intercalation compound SrSi<sub>2</sub>

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**Figure S1.** RHEED images along the [110] azimuth of the Si substrate. a) Reconstructed Si(111) surface. b) A film of SrSi<sub>2</sub> (d = 160 nm). The lattice match between the lateral lattice parameters of Si(111) and SrSi<sub>2</sub> is highlighted by red dashed lines.



**Figure S2.**  $\theta$ -2 $\theta$  XRD scans of ultrathin SrSi<sub>2</sub> films (the  $\theta$  angles are scaled to the Cu K<sub> $\alpha$ 1</sub> wavelength to facilitate comparison with Fig. 1c): a) 3 ML and b) 8 ML; asterisks mark peaks from the substrate.



**Figure S3.** Cross-sectional HAADF-STEM images of interfaces in  $SiO_x/SrSi_2/Si(111)$ : a) the  $SiO_x/SrSi_2$  interface; b) the  $SrSi_2/Si(111)$  interface. Both images are viewed along the [110] zone axis of the Si substrate.



**Figure S4.** Cross-sectional HAADF-STEM images of ultrathin SrSi<sub>2</sub> films on Si(111): a) 1 ML; b) 2 ML; c) 3 ML; d) 8 ML. The images are viewed along the [110] zone axis of the Si substrate. Bright points correspond to Sr atoms.



**Figure S5.** Electron transport in a thick  $SrSi_2$  film (d = 160 nm). a) Longitudinal and b) transverse conductance at 2 K in out-of-plane magnetic field; red lines provide fits of the data by a two-band model. Temperature dependence of c) carrier mobility and d) carrier concentration for the electron (red) and hole (blue) bands.



Figure S6. Electrical scheme dependence of magnetoresistance of a thick  $SrSi_2$  film (d = 160 nm) at 2 K in magnetic field parallel to the current: a) 2-terminal (red) vs. 4-terminal (blue); b) interchange of potential and current contacts.



**Figure S7.** Resistivity of a thick  $SrSi_2$  film (d = 160 nm) at T = 2 K (blue) and T = 5 K (red) in magnetic field parallel to the current.



**Figure S8.** Dependence of a) longitudinal and b) transverse resistivity in a thick  $SrSi_2$  film (d = 160 nm) at T = 2 K on the angle  $\phi$  between an out-of-plane magnetic field (with in-plane projection along the current direction) of 14 T and the current direction (blue dots) and their respective fits as  $const \cdot sin^2 \phi$  and  $const \cdot sin \phi$  (red lines).



**Figure S9.** Dependence of a) longitudinal and b) transverse conductivity in a thick  $SrSi_2$  film (d = 160 nm) at 40 K (blue) and 70 K (red) on the angle  $\theta$  between an in-plane magnetic field of 14 T and the current direction.



Figure S10. Magnetoconductance in 3 ML SrSi<sub>2</sub> at T = 2 K in magnetic field parallel to the current.



**Figure S11.** Temperature dependence of resistivity in SrSi<sub>2</sub> films of different thickness: 1 ML (red), 2 ML (cyan), 3 ML (orange), 8 ML (blue), 10 ML (green), 30 nm (magenta), and 140 nm (grey).



**Figure S12.** Magnetoresistance of 8 ML  $SrSi_2$  at T = 2 K in magnetic fields directed in-plane (parallel to the current, blue) and out-of-plane (red).



**Figure S13.** Magnetoresistance of 10 ML  $SrSi_2$  at T = 2 K in magnetic fields directed inplane (parallel to the current, blue) and out-of-plane (red).



**Figure S14.** Temperature dependence of a) carrier mobility and b) carrier concentration for electrons (red) and holes (blue) in 140 nm SrSi<sub>2</sub>.



**Figure S15.** Temperature dependence of a) carrier mobility and b) carrier concentration for electrons (red) and holes (blue) in 10 ML SrSi<sub>2</sub>.



Figure S16. Magnetoresistance of 30 nm  $SrSi_2$  at T = 2 K in in-plane magnetic field, parallel (blue) and orthogonal (green) to the current, and out-of-plane magnetic fields (red).



Figure S17. Magnetoresistance of a  $SrSi_2$  film (d = 30 nm) at T = 2 K in magnetic field parallel to the current.



**Figure S18.** Two-terminal measurements of I-V characteristics in a thick  $SrSi_2$  film (d = 160 nm) at 2 K (red), 20 K (green), and 50 K (blue) demonstrating an ohmic behavior. Inset shows a scheme of the measurements – the sample (blue) and the contact areas (grey); the distances are given in mm.