Supplementary Information: Atomic structures and interfacial engineering of ultrathin indium intercalated between graphene and SiC substrate

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Fig. S. 1. Calibration of STM tip using field emission regime (FER). (a) Schematic illustration of a STM junction at high bias voltage in FER regime yielding hydrogen-like resonances (red) confined in the vacuum region between the tip and sample. (b) FER spectra (dI/dV spectra with closed feedback loop) obtained on clean graphene/SiC showing series of resonance peaks.

To calibrate the tip (in situ) without changing the samples, we perform conductance spectroscopy in the field emission regime (FER) by applying high positive bias voltage between the tip and the sample (typically
between 2 V and 10 V). In the FER regime, the Fermi level of the tip is higher than the vacuum level of the sample. Thus, the electric field induced by the tip lifts up the potential barrier above the vacuum level of the sample, giving rise to a set of hydrogen-like states confined to the vacuum region between the tip and the sample. We repeatedly record two dI/dV spectra by ramping the bias forward from 2 V to 10 V and backward from 10 V to 2 V until two series of pronounced peaks in both voltage bias directions are well-observed with sharp resonance peaks without instability. We believe that the high electric field emitted from the tip helps somehow “re-form” the tip apex and remove the non-metallic dirt attached to the tip. This technique has been applied during our work and shown very high fidelity.

Fig. S. 2. (a) A SiC step edge with the height of 3 Å as shown in the height profile in (b).

We observe few step edges in our sample with the height profile of 3 Å. This step height is attributed to the original SiC step edge, however both regions shown here are also fully intercalated by indium (exhibiting smooth surface feature). This step edge is distinguished from the step edge shown in Fig. 1 (a) with the height of only ~ 1.2 Å which is formed due to different indium coverage on the same atomically flat SiC terrace.
In Fig. S. 3, the orange spectrum is taken on the newly formed higher region (brighter contrast, marked as orange dot in the STM image of the inset) which is formed after the voltage pulse. The black spectrum is taken on the lower region (marked as black dot in the inset). Both spectra show very similar features with initial spectra taken on the low and high regions in Fig. 1 (d). This indicates that upon tip manipulation, the newly formed region is identified as bilayer indium showing exactly the same electronic structures with that of the initial high intercalated region.